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Hydrogeology and Environmental Geology

Environmental History of Lake Hältingträsk and Recent Times in Lake Storträsk

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<p>Tiivistelmä/Referat – Abstract</p> <p>The European Water Framework Directive aims at restoring all water bodies in good ecological conditions by the year 2023. For this aim, understanding the responses of these ecosystems to current and future pressures is a requisite. Lakes Hältingträsk and Storträsk are located in Östersundom, a latent developing suburban area in eastern Helsinki. Alterations to the catchment in Hältingträsk as a consequence of urbanization will likely change the conditions of the lake. Storträsk, part of Sipoonkorpi nature reserve is primarily influenced by recreational activities. Ecological status of both lakes is likely to alter under the ongoing urban development. For this reason, the reference conditions of Hältingträsk and the resilience of both lakes to human stressors must be assessed.</p> <p>A long term record from Hältingträsk, with special focus on the most recent section, as well as a short core from Storträsk targeting the most recent events, are analyzed for different palaeobiological and geochemical proxies. The sequence from Hältingträsk is evaluated with diatom assemblages, trace metal analyses, lithological description of sediments through loss-on-ignition and inferred chlorophyll a. For Storträsk, a high-resolution study of diatom communities and photosynthetic pigments is performed. Both sequences are framed with an age-depth model based on radiogenic dating techniques. In addition, the results are analyzed with statistical tools and fossil diatom data is used to reconstruct lake water pH.</p> <p>The results describe the evolution of Hältingträsk through the mid-Holocene until recent times; the diatom assemblages indicates the area was part of Ancylus Lake and, later of Litorina Sea, and that it was isolated from the Baltic Basin at 6500 cal BP. This is supported by the high concentrations of Fe and Mn, showing the presence of metallic nodules common in marine environments. The change in sediments and the predominance of fragilarioid diatoms, display the succession of the lake (from gloe to flada). Afterwards, the ontogeny of the lake and the development of surrounding peat bog can be tracked with changes in the diatom community and decrease in heavy metals concentrations. The reconstructed pH reveals that Hältingträsk is a naturally acidic lake. Furthermore, signals of agricultural activities and industrialization are recorded in the area, as well as their development, is recorded through shifts in the diatom community and the oscillation of trace metals of both local (Cu, Ni and V) and long (Pb, Zn and Cd) transport. Finally, climatic anomalies such as the Little Ice Age and current climate warming are imprinted in the diatom assemblages and the photosynthetic pigments.</p> <p>The high resolution of subsampling from Storträsk displayed little variation. The faint changes could be attributed to CaCO<sub>3</sub> treatment, fish introduction or recent climate warming. However, discern the influence of each of these stressors was not possible.</p>			
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## 1. INTRODUCTION

### 1.1 Stories Told by the Bottom of Lakes

It is a well-known fact that lake ecosystems are affected by modifications to their catchment, air pollution and climate change on top of their own ecosystem succession (Virkanen et al., 1997, Korhola et al., 2000, Smol et al., 2005, Schindler, 2009, Fritz and Anderson, 2013). Owing to the lack of numerical records extending far back in time, it is not possible to understand the natural variability and the evolution of the ecosystems existing nowadays. In addition, the environments currently change at a faster speed than they would without anthropogenic stressors (Battarbee and Bennion, 2012). Lakes, especially those in northern latitudes, are sensitive to modifications (Korhola et al., 2000, Smol et al., 2005, Weckström et al., 2006). Thus, the need to study the past lake-conditions arises.

One way to decipher the evolution of ecosystems is through natural archives. The horizontal homogeneity of lake deposits makes them good environments to study the past, as the sediments at the bottom represent the conditions of the entire catchment (Schindler, 2009, Smol and Stoermer, 2010). The signals imprinted in the sediments are not limited to the local effects and the internal dynamics of the lake, they also archive traces of global change (Smol et al., 2005, Schindler, 2009, Battarbee and Bennion, 2012). The changes registered can be long-term variations such as the subdivisions of the Holocene, with the warm and dry period between 8200 and 4200 BP (Walker et al., 2012), or climate anomalies of the late Holocene, for instance the Medieval Warm Period and the Little Ice Age (Tiljander et al., 2003). Likewise, it is possible to track changes in shorter periods of time with high resolution studies. This enables to distinguish signs of anthropogenic impact in the environment, such as atmospheric pollution or human induce climate change (Virkanen et al., 1997, Hakala and Salonen, 2004, Battarbee and Bennion, 2011).

Palaeolimnology studies the sediments of waterbodies in order to reconstruct the past. Some of the evidence found in lakes are fossil biota such as diatoms, chemical residues from atmospheric and hydrological input, and residues of the trophic dynamics; photosynthetic pigments or organic carbon for example. The development of this science

goes hand in hand with simultaneous efforts to protect and restore lakes (Battarbee and Bennion, 2011). In Europe, the Water Framework Directive states that all water bodies should be in good status by 2027 (European Union, 2000). In this context, good ecological conditions were defined as having only slightly deviated levels from the undisturbed environment, regarding the biological and chemical values (European Union, 2000). With a view of recovery and conservancy, it is necessary to comprehend the natural state of the environment before human influence and establish reference conditions. An evaluation and comparison of the current state with past conditions is the key to assess the resilience of lakes to ongoing global change.

The EU has aimed at preserving, protecting and restoring freshwater bodies to good ecological condition (European Union, 2000, Deutsch et al., 2013). Lakes in southern Finland, the most populated region of the country, are exposed to direct human activities and therefore are a matter of discussion and study. Lakes Hältingträsk and Storträsk are located in the municipality of Helsinki where, as seen in Figure 1, lakes have generally poor ecological status (National Land Survey of Finland, 2013). Furthermore, the sub-district of Östersundom, where the lakes are, is under development. The assessment of the evolution of lakes in Sipoo has been done before (Sarmaja-Korjonen, 1992, Luoto, 2009, Rantala, 2013, Pellikka, 2018). The general appraisal is that lakes in the area were isolated from the Baltic Basin during the Ancylus Lake stage or the Litorina Sea stage, and after a period of transition they evolved into small acidic humic lakes surrounded by *Sphagnum* bogs (Sarmaja-Korjonen, 1992, Seppä et al., 2000a, Luoto, 2009, Rantala, 2013). Sarmaja-Korjonen (1992) states human settlements began near Hältingträsk in 2800 BP (3000 cal BP), and both Luoto (2009) and Rantala (2013) demonstrates the sensitivity of lakes Storträsk and Hampträsk to human stressors. Because of the future urban growth in the area, understanding the evolution of the lakes is a requirement. The terrain near Storträsk is in the nature reserve of Sipoonkorpi and therefore cannot be modified (Pellikka, 2018). But before further alterations take place, it is preponderant to define the reference conditions of Hältingträsk and vulnerability of both lakes.

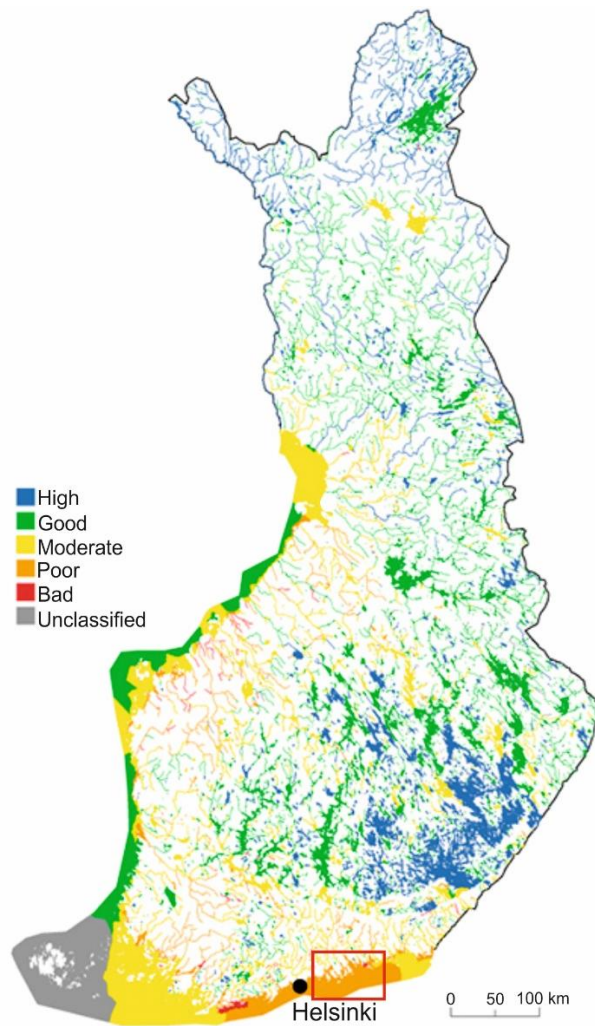


Figure 1. Ecological status of the surface waters of Finland in 2013. Southern Finland waterbodies have badly to moderate ecological status. The study area reports poor ecological status in general and is highlighted with a red rectangle. After the Assessment of the ecological status of Finland's surface waters (National Land Survey of Finland, 2013).

## 1.2 Objectives and Hypothesis

The general objective is to establish the reference conditions of Hältingträsk, considering the ecological responses to human pressures associated with urban expansion, in particular with regards to the future urban development in the catchment of the lake. Associated with this aim and in favor of proper water management, specific goals are set; to assess resilience to anthropogenic influences and the atmospheric input in Hältingträsk, as well as its recent trophic development through the geochemistry, analysis of the productivity and fossil biota in the sediments. This study also aims to describe the evolution of the Lake Hältingträsk, its development through the mid-Holocene until the present days, as well as detect signals of past human influence in the sedimentary records

with special focus on the recent history. To achieve these goals, a reconstruction of environmental variables based on diatom assemblages along with physical and chemical analysis of the sediments were framed with an age-depth model.

Regarding Storträsk, the objective is to evaluate the effects of the recent fish introductions, the liming treatment and the possible affectations to the ecological status in recent years. For this study, with focus on recent changes, diatom assemblages and photosynthetic pigments are used to analyze possible changes in the catchment.

The evolution of the lakes Hältingträsk and Storträsk is imprinted in the geochemistry and fossil record of the sedimentary column. The current state of the lake Hältingträsk with acidic water and lack of fish, as well as the poor or inexistent buffer capacity of both lakes, is a result of their evolution. Moreover, the modifications to the catchment, consequence of human activities close by, for example agriculture and ditching, have as well modified the lakes. The biological (diatoms) and physicochemical (trace elements, pigments and organic matter content) properties of the sediments are expected to reflect these processes, therefore providing tools to disclose the history of the lakes.

## **2. STUDY SITE**

Lakes Storträsk and Hältingträsk are located in southern Finland in Ostersundom area, a territory that used to belong to Sipoo municipality but now is part of eastern Helsinki (Fig. 2). The city planning contemplates future urban development near the catchment of the lakes, in addition to the ongoing recreational activities and agriculture (Pellikka, 2018). The catchment of Storträsk is in Sipoonkorpi nature reserve, consequently it will not be subject of construction. However 63% of the catchment of Hältingträsk is expected to become an urban district (Pellikka, 2018). The modifications of the area are likely to change the ecological status of the lakes.

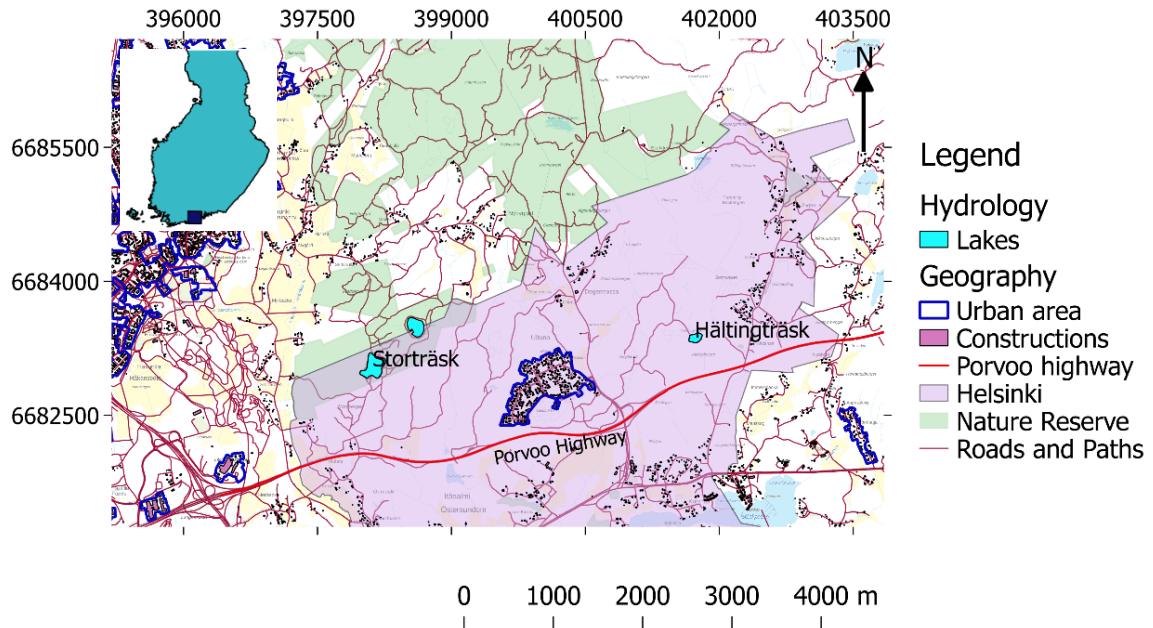


Figure 2. Location of the two lakes inside the Helsinki Municipality. The area of Helsinki displayed in the map is Östersundom, a neighborhood with plans for urban development. The map also indicates the protected area where Storträsk is in, as well as the location of the study site in Finland (National Land Survey of Finland, 2014).

## 2.1 Geology and Recent History of the Lakes

Both lakes are situated close to the shore of the Baltic Sea, Storträsk a couple of kilometers and Hältingträsk around 5 km away. Here, it is common to find lakes whose formation is the result of the land uplift after the deglaciation of the Fennoscandian Ice Sheet in southern Finland, starting in 12700 cal BP, and ending in 9700 cal BP (Miettinen, 2004, Stroeve et al., 2016). The Ancylus Lake (10300 to 8200 cal BP) became brackish on its late phase, after the sea level rose as a result of isostatic adjustment and the Danish Straits became flooded, allowing an exchange between the Baltic Basin and the sea (Miettinen, 2004, Björck et al., 2008). The following transition towards a more saline environment is known as Litorina Sea, and started around 8200 cal BP (Miettinen, 2004). The isolation of lakes from the Baltic Basin followed a succession from glo lake with slightly brackish water to a flada, a fresh-water lake isolated by isostatic uplift during this period (Seppä and Tikkanen, 1998, Miettinen et al., 1999, Seppä et al., 2000b, Miettinen, 2004). Furthermore, in southern Finland, the ancient shoreline of the Litorina Sea has its highest points between 20 and 40 m a.s.l, placing both lakes in the area of influence of this stage (Ojala et al., 2013). The altitude of Hältingträsk (28 m a.s.l.) and Storträsk (31 m a.s.l.) can be seen in the map from Figure 3. Also, past analysis suggests the isolation of Hältingträsk happened at around 6000 BP (6500 cal BP) (Sarmaja-Korjonen, 1992).



The geology observed in the area nowadays consists of elevated, exposed and fractured bedrock, with quartz-feldspart gneiss, microcline granite and quartz granodiorite from the Svecofennian period (1930-1780 Ma) (GTK, 2018). The bedrock surrounding the lakes has enclosing valleys as predominant topographic features to the east and west. The narrow and long valleys are faults filled with till and peat deposits while the fractures associated to the faults are filled with fine sediments and clay (GTK, 2012, GTK, 2018). Figures 4 and 5 show the principal surface deposits surrounding the lakes, and the catchment of Hältingträsk and Storträsk.

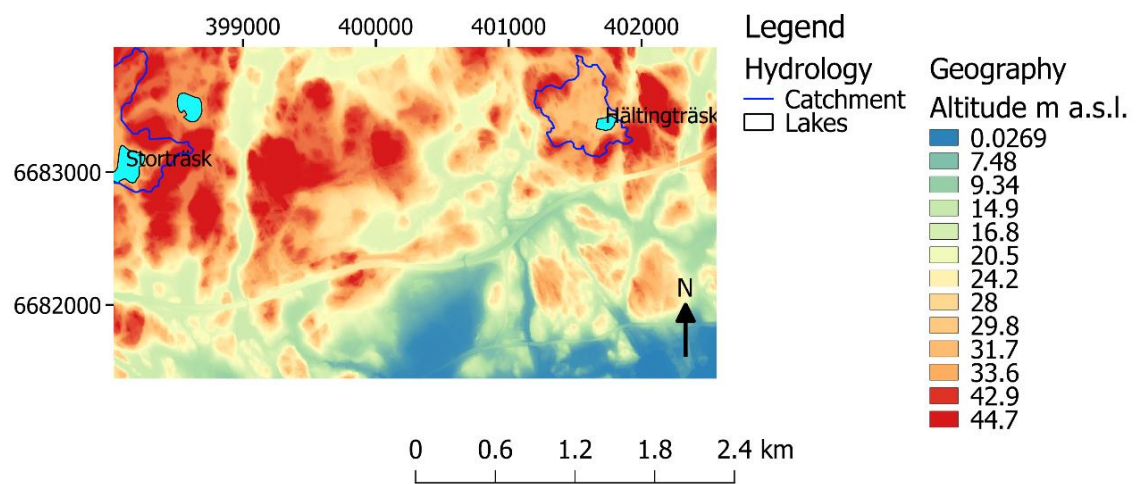


Figure 3. Digital elevation model based on Lidar imaging, in the map both lakes are displayed. Hältingträsk has an altitude of 28 m a.s.l. and Storträsk of 31 m a.s.l. (National Land Survey of Finland, 2016).

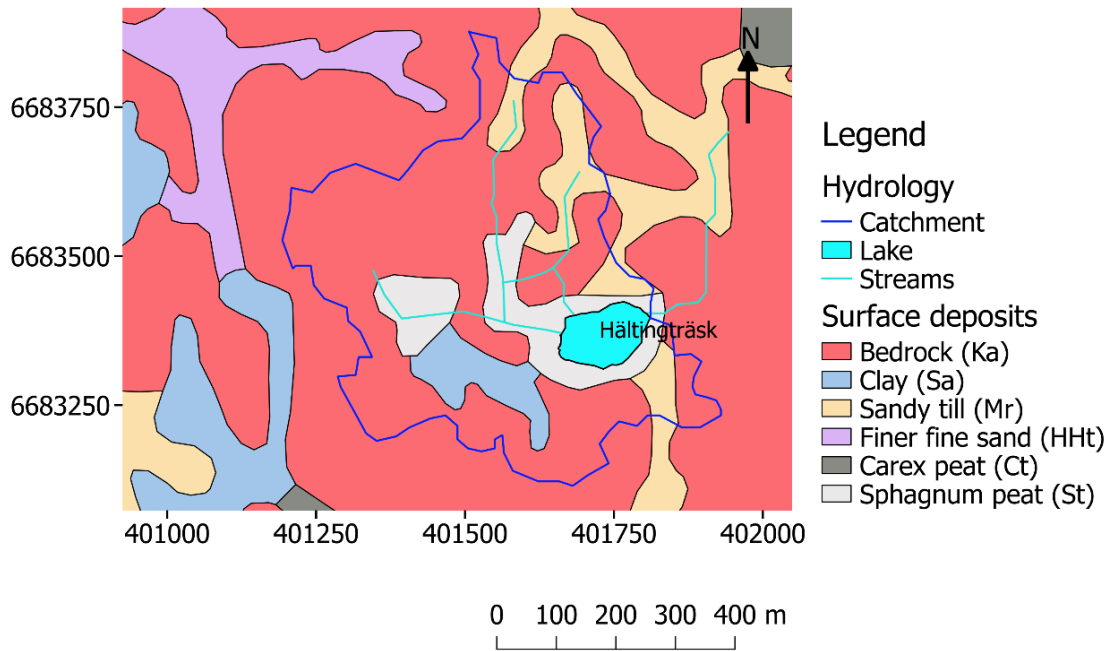


Figure 4. Lake Hältingträsk and the surface deposits found in the catchment, the lake is surrounded by *Sphagnum*-bog, it also has clay and sandy moraine till deposits, and exposed bedrock. Map after the Bedrock of Finland DigiKP200 (GTK, 2018).

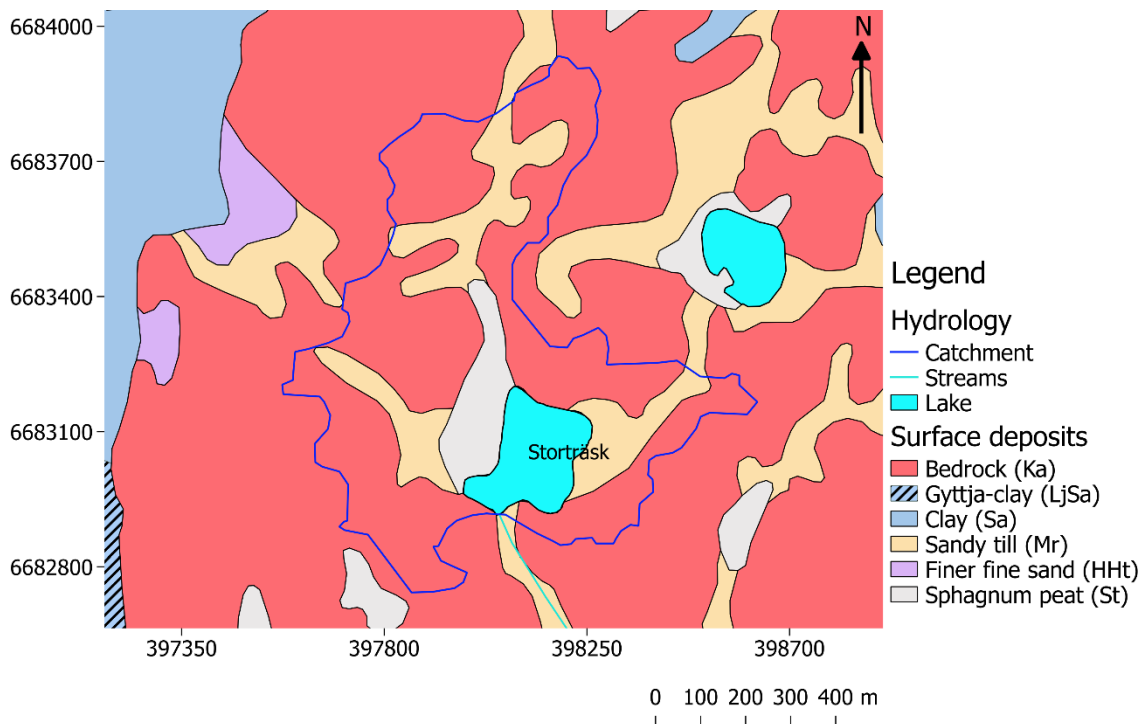


Figure 5. The main surface deposits near Storträsk are *Sphagnum*-bog, sandy moraine deposits and exposed bedrock. Map after the Bedrock of Finland DigiKP200 (GTK, 2018).

As the area was rising from the Baltic Basin, no human settlement occurred at that time, the seldom stray finds point to a sparse occupation until the Stone Age and the Early Bronze Age. Thus, activities like fishing and hunting did not transform the local

vegetation (Sarmaja-Korjonen, 1992). The archeological findings near Hältingträsk, about 1.5 km to the south, imply the population began to grow in the Late Bronze Age and the Pre-Roman Iron Age. Nevertheless, it was not until 1200 BP (1300 cal BP) that agriculture began, prevailing until present time in the vicinity of the catchments (Sarmaja-Korjonen, 1992). Other human activity around the lakes, is the recreational use, for instance in Storträsk fish plantings are a common practice with species such as white fish (*Coregonus lavaretus*) and trout (*Salmo trutta*) (Kujala, 2011, Pellikka, 2018). In this regard, Storträsk has received liming treatment to improve the conditions for the fish, however it is not known when both practices started (Pellikka, 2018). Southern Finland is the most populated area in the country, and in consequence urbanization also has an influence on the lakes. Structures like the highway Porvoonväylä are a source of atmospheric chemical input in the catchment as well (Verta et al., 1989, Virkanen et al., 1997, Skjelkvåle et al., 2001, Rantala, 2013).

## 2.2 Ecology and Hydrology of the Lakes

The catchment of Storträsk is small; around 56.4 ha for Storträsk with ~4.5 ha in lake surface and a maximum depth of 5 m. Whereas the smaller lake Hältingträsk has a catchment close to 30 ha, a surface of 1.29 ha and a maximum depth of 4 m (GTK, 2012, Pellikka, 2018). In the surroundings of the shore, both lakes have *Sphagnum*-type peat and boreal forest with pine trees, Storträsk has bog vegetation only in the north-western margin, while Hältingträsk is completely enclosed by it (Figures 4 and 5). The hydrology in Storträsk is governed by precipitation and seeping as inputs and a small channel as output, conversely, the peat surrounding Hältingträsk has ditches that serve as additional water inlets (Pellikka, 2018). The climate in the municipality of Helsinki is humid and continental, the average temperature during summer is 16°C, and -3.5°C in winter. Since the decade of 1990 the records report positive anomalies, implying a warmer climate in the past thirty years and shorter winters (Korhonen, 2006, Finnish Meteorological Institute, 2019).

Previous studies in Storträsk (Rantala, 2013) show that the water pH is acidic by nature and has little buffering capacity, but CaCO<sub>3</sub> treatments over recent years affect the alkalinity of the water. On the other hand, Hältingträsk has not undergone liming and has no buffering capacity. The lakes are eutrophied to some extent, according to their nutrient

measurements in the last decade (Table 1). Regardless of the general ecological status in the region (Fig. 1), Storträsk was determined to be in good ecological condition and Hältingträsk in excellent condition (Pellikka, 2018).

Table 1. Geographic and ecologic information of lakes Storträsk and Hältingträsk. The elevations have been calculated from DEM from Lidar imaging (Fig. 3) (National Land Survey of Finland, 2016), and the water quality measurements correspond to the monitoring data from 2012 to 2017, the data is part of the City of Helsinki report (Pellikka, 2018).

	Storträsk	Hältingträsk
Coordinates	N 6683036.9 E398119.6	N 6683361.4 E 401725.9
Catchment area (ha)	56.4	29.58
Lakes surface (ha)	4.49	1.29
Elevation (m)	31	28
Maximum depth (m)	5	4
pH	4.5 to 6	4 to 6
Water oxygen content (mg L <sup>-1</sup> )	0 to 14	0 to 12
Water alkalinity (mmol L <sup>-1</sup> )	0 to 0.2	-0.05 to 0.05
Total nitrogen content (µg L <sup>-1</sup> )	400 to 800	400 to 1200
Total phosphorus content	5 to 30	0 to 40
Nitrogen from Ammonium, nitrates and nitrites (µg L <sup>-1</sup> )	0 to 300	0 to 300
A-chlorophyll content on surface water	0 to 40	0 to 30

### 3. MATERIALS

The sediments for the study come from cores collected on two different occasions; the first field campaign took place in early March 2018 and the second in late August 2018. In the winter, a short core was taken with an HTH gravity corer from both lakes. The 3.5 cm sediment core from Storträsk was subsampled with a 2.5 mm resolution, while the 18.5 cm core from Hältingträsk was subsampled at a 2.5 mm resolution for the top 5 cm and 5 mm resolution for the rest. The samples were collected in sealable bags. An additional long core of 405 cm was taken with a Russian peat corer from Hältingträsk in winter, with the cores stored in plastic tubes. All sediment samples were stored in a cold room at 4°C afterwards. The subsampling of the long core was done according to its purpose; for loss-on-ignition the sediments were subsampled at a 5 cm resolution, for trace metals the first 1.5 m had the same 5 cm resolution, followed by a 10 cm resolution for the remaining of the sequence, and finally, for diatom analysis the core was subsampled at a 1 cm resolution. However, for the diatom analysis, of the first 1.25 m only samples every 10 cm were used and for the rest of the sequence only the samples

every 20 cm were analyzed. Depending on the analysis the samples were stored in different manner. For determination of heavy metals concentrations and diatom analysis the samples were freeze-dried and for LOI the sediments were kept frozen and were thawed afterwards for their measurements. The summer field work consisted of taking complementary short cores of 12 cm subsampled at a 5 mm resolution with HTH gravity corer from both lakes. The purpose of these samples was to measure the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  activity for dating. For this reason, the sediments were placed in sealable bags that were previously weighted to measure their water content.

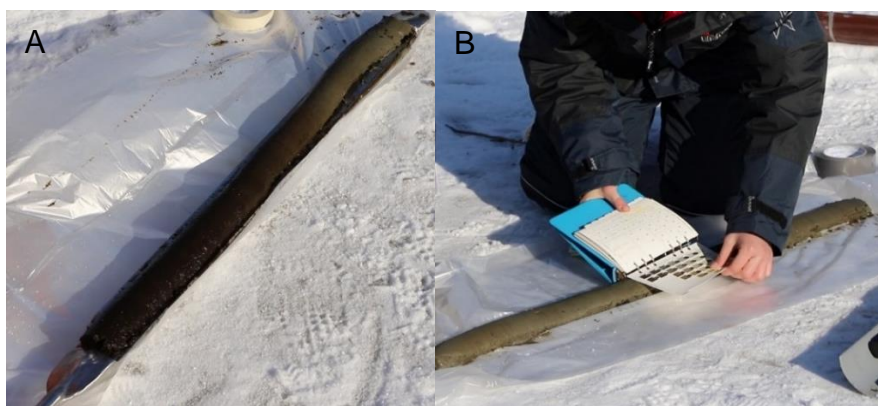


Figure 6. Sediment cores from Hältingträsk taken with a Russian peat corer. The picture A shows the transition between sediments with high organic content and sediments from the gyttja type, while picture B displays one of the bottom-most cores with clay and fine sediments. In the same picture B, it can be seen the color analyses with the Munsell Soil Color Chart.

## 4. METHODS

### 4.1 Dating Methods

To establish the chronology for the sequences, the sediments were dated with radiogenic dating techniques. The most surficial sediments, from the two cores taken in summer, were dated with  $^{210}\text{Pb}$ . This method was chosen for it is more suitable for recent sediments with ages ranging from 0 to 200 years old (Krishnaswamy et al., 1971). Additionally,  $^{137}\text{Cs}$  was also measured to mark the nuclear bomb testing and the Chernobyl nuclear accident (Klaminder et al., 2012). The measurements were made in the Paleoecological Environmental Assessment and Research Lab (PEARL) in Canada.

For the long sequence in Hältingträsk, four macrofossils at depths with a peculiar signal in the sediment geochemistry (48 cm, 109 cm, 173 cm and 302 cm) were selected to be dated by the  $^{14}\text{C}$  method with an Accelerator Mass Spectrometer (AMS). The

measurements were done by the Radiochronology Laboratory C.E.N in Canada. Using plant macrofossils was preferred over bulk sediment to avoid the reservoir effect (Geyh et al., 1997), which can yield results influenced by the different sources of C in the basin. Finally, in order to generate an age-depth model, the radiocarbon dates were calibrated and interpolated using the R based code Clam for calibrating and modeling (Blaauw, 2010). The model used was smooth spline with a smooth factor of 0.4, with 95% of confidence ranges and 10000 iterations, this model was chosen because it gave the best fit. The calibration curve was IntCal13, as it is recommended for the northern hemisphere (Reimer et al., 2013). In addition, a 400 year reservoir correction was performed in the sample from the depth 173 cm, a typical correction for sediments from Litorina Sea (Krog and Tauber, 1974, Rößler et al., 2011, Bendixen et al., 2017). Additionally, uncalibrated dates referenced from literature were calibrated as individual dates using the IntCal13 curve and the software Oxcal for better comparability with the current results (Ramsey, 1995)

## **4.2 Lithological description**

When the long cores were taken during the winter field campaign, they were described using the Munsell Soil Chart to capture the fresh colors of the sediments (Fig.6). Afterwards, loss-on-ignition (LOI) analysis was performed with the LECO TGA701 Thermogravimetric Analyzer at the Environmental Laboratory in the University of Helsinki, following the SFS 3008 standardized method for measuring moisture and LOI at 550°C this procedure is based on the original methodology (Dean, 1974). In general, clean dry crucibles are weighted before the placement of the sample, then with the wet sample, and finally with the burnt dry sample. In every 20 samples analyzed, at least one blank crucible, a duplicate sample and one crucible with reference material (WQB-1) were included. Loss-on-ignition is a common control method to estimate the organic matter and carbonate content in sediments (Heiri et al., 2001), as wells as to make inferences on the conditions of the catchment (Mohr et al., 2000, Heiri et al., 2001). The samples taken during the summer were used to measure the water content by weighting the Minigrip bag on its own, then with wet sediment, and finally the sealable bag with the dry sediment after freeze-drying.

## **4.3 Diatom Analysis**

To investigate trends within the lake, in the surrounding catchment and the climate, fossil diatoms were analyzed in both sediment sequences. The microscope slides were prepared with dilutions made from the freeze-dried sediments, the samples were oxidized with hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) and heated to  $55^\circ\text{C}$  to remove organic matter, as the standard procedure indicates (Battarbee and Kneen, 1982, Berglund, 1986, Renberg, 1990). After repeated decanting and washing of the samples, a small amount of the diatom suspension was placed on the slides, then, after the dilution was dry the covers were mounted with Naphrax®. The diatom identification and counting were done with a light microscope at 1000x magnification, and least 300 diatom frustules were counted per sample. As for the reference and guides for the diatom identification, the main source was the series of books for the flora of freshwater of Central Europe (Krammer and Lange-Bertalot, 1986-1991).

#### **4.4 ICP-MS Geochemistry Analysis**

The sediments from Storträsk were already analyzed for their trace elements concentrations previously (Rantala, 2013), therefore only the geochemistry of Hätingträsk was examined. The concentrations were measured by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) in the Agilent 7900 ICP-MS at the Environmental Laboratory of the University of Helsinki after being digested in the microwave oven Cem Mars5. First, an approximate amount of 0.25 g of freeze-dried sample was weighted into Teflon tubes, then 10 ml of  $\text{HNO}_3$  was added to the sample for its digestion in the microwave oven, afterwards, the sample was diluted 20 times in Milli-Q® water prior the measurement. The elements measured were phosphorus (P), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), cadmium (Cd) and lead (Pb). To calibrate the spectrometer a semi quantitative analysis was performed first with 20 samples from different depths but covering the whole core.

To control the quality of the results, one blank and one sample duplicate every 16 sample were included in the microwave oven. Moreover, two different reference materials (WQB-1 and LKSD-4) were measured with duplicates for each. Also, based on the semi quantitative analysis, six standard solutions were prepared into 10 ml vials with declining concentrations, the stock solution was made with certified materials of known concentrations. To normalize the results and as control method to identify errors and instrument drift, a series of internal standards were used, and the correlation coefficients

of the calibration curves were observed. Ultimately, the concentrations in the sediments were calculated with a blank-correction.

#### **4.5 Inferred Chlorophyll a**

The sediments from the summer campaign had their levels of chlorophyll a measured by Visible Range Spectroscopy (VRS at the Paleoecological Environmental Assessment and Research Lab (PEARL) in Canada as described in Michelutti and Smol (2016). The levels of chlorophyll a and its degradation products (chlorins), are used as an indicator of trophic level in lakes. Considering an aquatic environment enriched in nutrients will have a higher production, the photosynthetic pigments will be higher than in oligotrophic lakes in consequence (Michelutti and Smol, 2016).

#### **4.6 Data Analysis**

##### *4.6.1 Reconstructed pH*

Modern measurements of environmental variables in Hålingträsk have only existed for a small range of time (Pellikka, 2018). In the absence of such data, palaeoenvironmental reconstructions based on fossil proxies provide a tool to infer the environmental conditions of the past (Battarbee and Bennion, 2012, Juggins, 2013, Birks et al., 2014). In this study, a reconstruction of water-lake pH was performed using a transfer function on fossil diatom assemblages using the software C2 (Juggins, 2014). As it is accepted this biota is sensitive to changes in the pH (Birks et al., 1990). The training set used was from the European Diatom Database EDDI (Battarbee et al., 2000), including lakes from Norway, Sweden, Finland, England, and Russia. The reason to choose this training set was its wide range of sites and pH values.

The method used to perform the reconstruction was Weighted Averaging Partial Least Squares regression (WA-PLS). This numerical method has been tested and shown good results for data with short gradients like the one in Storträsk. Furthermore, it yields better results than WA for datasets with larger gradients, thus it is a robust method for compositional data (Ter Braak, 1993). Also, as a cross validation method, the model used bootstrapping with 100 cycles. Finally, the assessment of the reliability of the reconstruction and representativeness of the training set over the fossil taxa was done with



the Modern Analogue Technique (MAT). A cut-off value of 5 percentiles in the distance to the closest analogue was considered a good estimation, while 10 percentiles was a sign of a good representation of the trends. Other values employed to evaluate the robustness of the model were a coefficient of determination ( $R^2$ ) above 0.8, a root mean squared error of prediction (RMSEP) below 0.4 pH units and at least 50% of species with a relative abundance above 0.05% considered in the reconstruction.

#### 4.6.2 Statistical Analysis

Along with a qualitative analysis of the results, quantitative analysis was performed on the data to better understand and describe the variations and trends. These methodologies were carried out for both sediment sequences, but due to small variation in the diatom assemblages from Storträsk, only the results from Hältingträsk are presented unless specified. The primary statistical methods applied on the data were cluster analysis to heavy metal concentrations and the diatom fossil data, as well as unconstrained ordination analyses on the diatom assemblages. For the cluster analysis, a hierarchical classification following the centroid clustering method (k-means) with squared Euclidian Distance as a measure of proximity was used. For the trace elements the values were standardized by their Z-scores whereas the diatoms, already expressed in relative abundance, were not transformed. The analysis was carried out using the software SPSS Statistics (IBM, 2016).

To explain the variability of the diatom taxa, unconstrained ordination was executed on the software CANOCO 5 (ter Braak, 2014). The compositional data was first subject of detrended correspondence analysis (DCA), to test the size of the variability gradient and determine if a linear or unimodal ordination was viable. Considering that the whole record in Hältingträsk is unimodal with a gradient of 6 SD linear ordination was dismissed. For this reason, principal component analysis (PCA) was only performed on the top samples with high resolution, which had a smaller gradient. Because the diatom taxa had a high abundance of rare taxa, the ordination was down weighted for rare environmental variables.

The assemblages of both cores showed a high taxonomic diversity of diatom species. Therefore, to assess species richness and evenness through the sequences two diversity indices were used; the number of taxa to assess the species richness, and Hills N2 for the

species evenness. The indices were calculated and analyzed with the software C2 (Juggins, 2014).

## 5. RESULTS

### 5.1 Age-Depth Model

The age-depth model framing the sediment sequence from Hältingträsk is based on calibrated radiocarbon dates, as well as  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  activity. The Table 2 summarizes the calibrated  $^{14}\text{C}$  ages, while the Figure 7 displays the final model and the activity of both Pb and Cs in Hältingträsk. The extrapolation model suggests an age of 8600 cal BP for the bottom of the sequence and the model did not find age reversals after the reservoir correction in the sample from 173 cm depth. Between the depths 109 cm and 48 cm, the model indicates an increment of accumulation rate, which translates into a flattening of the curve. Continuing, the final part of the model uses dates based on the activity of  $^{210}\text{Pb}$  and constrains the model with the  $^{137}\text{Cs}$  peak due to the Chernobyl accident of 1986 (Fig. 7). Both the Pb and Cs activity are in agreement in Hältingträsk, however, the samples reported high levels of Cs even before the nuclear disaster of 1986, this could be due to migration of the element or the nuclear bomb testing during that time.

Table 2. Depth of the macrofossil analyzed, age of the samples after the AMS analysis, and calibrated ages from the Clam age-depth model.

Depth (cm)	Lab ID	$^{14}\text{C}$ age (BP)	Modeled age (cal BP)		Best (cal BP)
			from	to	
48	ULA-8223	1715±15	1690	1567	1624
109	ULA-8226	4840±20	5606	5490	5578
173	ULA-8225	6520±20	7146	6939	7015
302	ULA-8224	7150±20	8008	7943	7973

In order to reach background levels in Storträsk, the summer core was longer than the winter sequence analyzed for diatoms. Unexpectedly, the background level were not reached within the 12 cm examined, and therefore the dates from Storträsk should be considered with caution. This can be seen comparing the activity of  $^{210}\text{Pb}$  in the two lakes (Figures 7 and 8), while Hältingträsk has low values comparable to  $^{214}\text{Pb}$  at the bottom,

Storträsk has activity levels above 5000 (Bq/kg) throughout the entire core. In a similar fashion as in Hältingträsk, the  $^{137}\text{Cs}$  activity was high before the maximum.

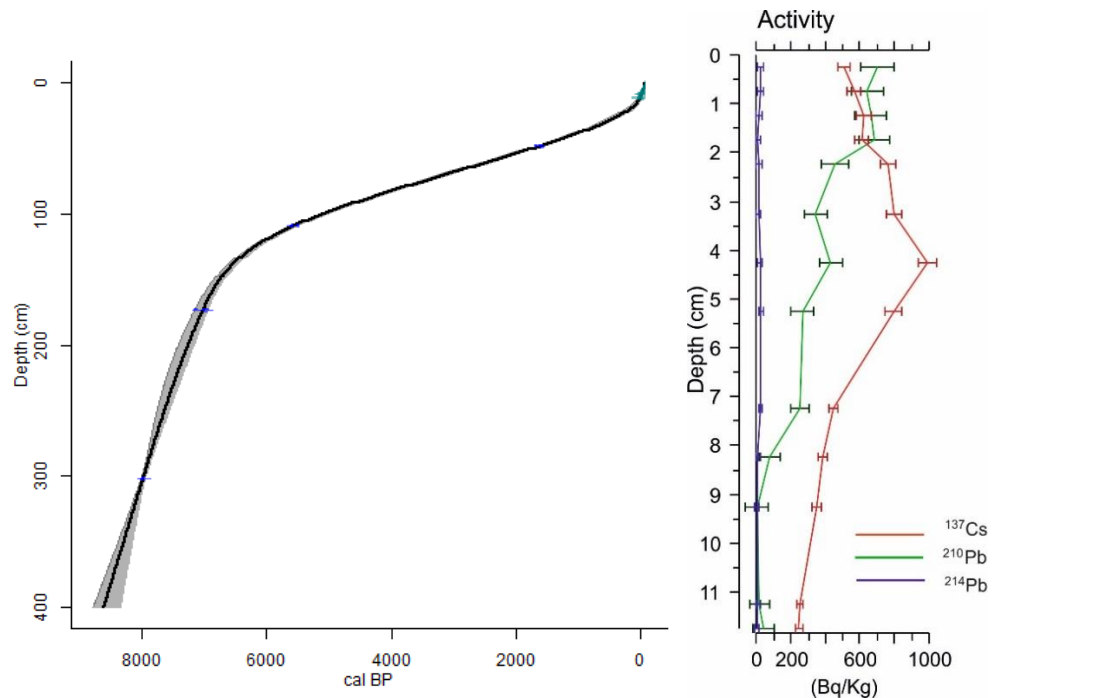


Figure 7. On the left, the age-depth model of the sediment sequence from Hältingträsk. The model extrapolated over the last centimeters of the sequence. On the right, the activity of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$ .  $^{210}\text{Pb}$  has background levels after 9 cm, and the nuclear accident in Chernobyl (1986) is detected around 4 cm depth.  $^{137}\text{Cs}$  shows high values even before this depth, perhaps due to Cs migration or enrichment after nuclear bomb testing.

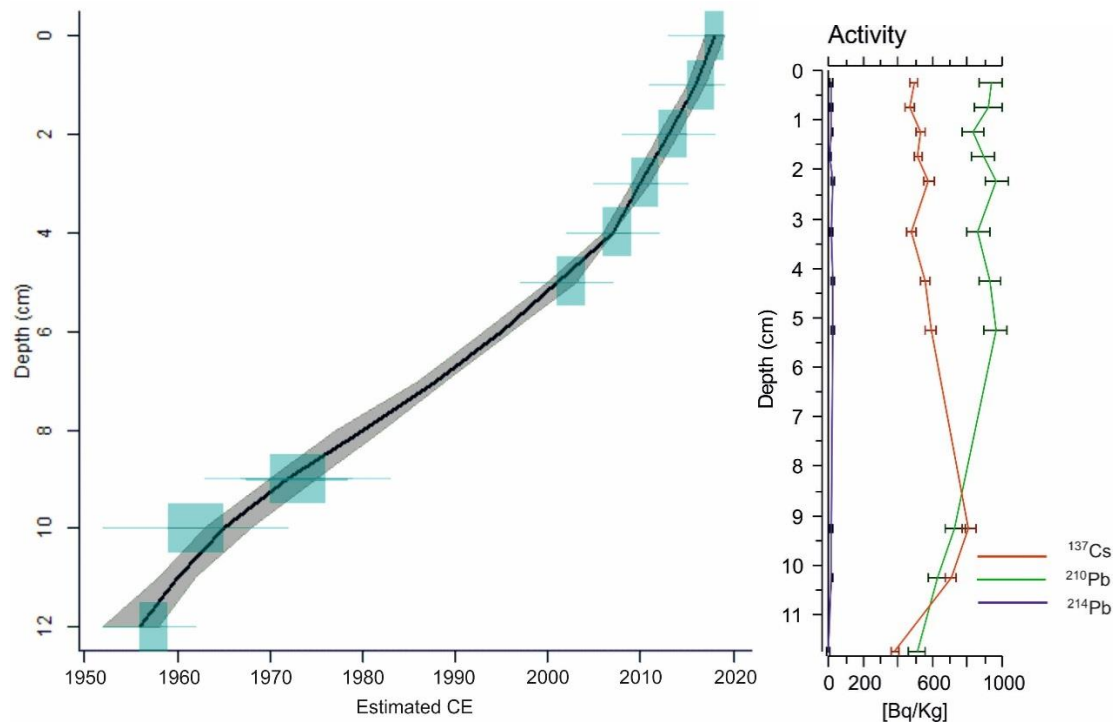


Figure 8. The age-depth model on the left corresponds to the sediments from Storträsk. As the activity levels on the right indicate, these results should be taken with caution, because Pb sequence did not reach background levels.

## 5.2 Lithology

The descriptions outlined in the Table 3 are from the sediments taken with the Russian peat corer. The color and the general features were reported in the field with the fresh sediments, while the rest of the data comes from analysis in laboratories. The sediments fall into four main categories according to their percentage of loss-on-ignition; Clay-Clayey-Gyttja (6-15%), Gyttja (15-40%), Dy (40-60%) and High-organic Gyttja (60-95%) after the classification of lake sediments (Salonen et al., 2002). There is a slight division in the clay-mud sediments that is not distinguished with the classification, from 405 cm to 305 cm the LOI is close to 10%, while from 305 cm to 215 cm the LOI is consistently about 12% (Fig. 9). Afterwards, the sediments report increasing percentages of organic carbon, first with ~70 cm of mud and finally with Dy. As the reported values of LOI were higher, accordingly the sediments had more macrofossils and higher water content.

Table 3. General characteristics of the sediments from the long core from Hältingträsk.

Core	Depth interval (cm)	Color (Munsell scale)	Description	Macrofossil sample for <sup>14</sup> C dating
C1-C2	0-95	Very dark brown 10 YR 2/2	<i>Features:</i> Abundant in organic remains. <i>LOI:</i> 62-84% - High-organic Gyttja	<i>Depth:</i> 48 cm <i>Age:</i> 1715 ± 15 BP <i>Age:</i> 1624 cal BP
C2-C3	95-145	Very dark brown 10 YR 2/2	<i>Features:</i> Abundant in organic remains, some stratification towards the bottom of C3. <i>LOI:</i> 55-63% - Dy	<i>Depth:</i> 109 cm <i>Age:</i> 4840±20 BP <i>Age:</i> 5578 cal BP
C3-C4	145-190	Very dark brown 10 YR 2/2 Olive grey 5Y 4/2	<i>Features:</i> Transition zone with black stripes (possible sulfide layers), less macrofossils. <i>LOI:</i> 20-55% - Gyttja to Dy	<i>Depth:</i> 173 cm <i>Age:</i> 6520±20 BP <i>Age:</i> 7015 cal BP
C4-C5	190-305	Olive grey 5Y 4/2 Dark grey 5Y 4/1	<i>Features:</i> stratification and bands, change to gyttja type sediments. <i>LOI:</i> 10-17% - Clayey-Gyttja	<i>Depth:</i> 302 cm <i>Age:</i> 7150±20 BP <i>Age:</i> 7973 cal BP
C6	305-405	Dark grey 5Y 4/1 Olive grey 5Y 4/2	<i>Features:</i> stratification and a few black bands (possibly charcoal). <i>LOI:</i> ~10% - Clayey-Gyttja	

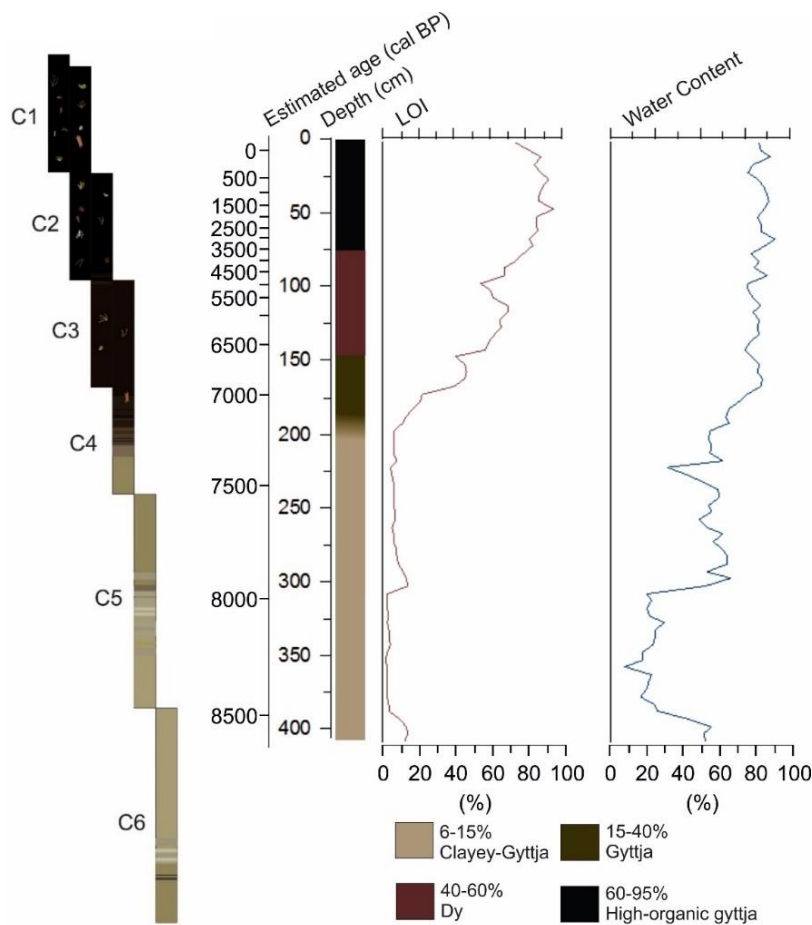


Figure 9. On the left, the figure displays the sequence of cores extracted with the Russian Peat corer with some elements identified in the field, such as high organic matter, stratification or black bands. The graph on the right has the percentages of LOI and water content, as well as a stratigraphic column with the classified sediments.

### 5.3 Diatom Stratigraphy

In the core from Hältingträsk 339 diatoms were identified to species level, but only 13 of them have abundances above 1% (Fig. 10). The most common species are represented in Fig 10. The araphid *Asterionella formosa* (26%) and *Tabellaria flocculosa* (17.6%) are the most abundant species in the record. Other pennate species with high abundance are from the genera *Eunotia* and *Pinnularia*, for instance *E. incisa* (6.5%), *E. bilunaris* (3.5%), *E. serra* (3.3%), *E. rhomboidea* (1.74%), *P. gibba* (1.4%), *P. subcapitata* (1.3%), and *P. microstauron* (1%), meanwhile *Aulacoseira tenella* was the only centric species with high abundance (4.9%).

There are three main shifts in the diatom species. The section from 405 cm to 220 cm is defined by a wider diversity of genera and species of diatoms from benthic environments, for instance *Fallacia*, *Amphora*, *Karayevia* and *Mastogloia*. Then, the second segment is

a transitional zone from approximately 220 cm to 160 cm that displays a higher abundance of *Staurosira*, *Pseudostaurosira* and *Staurosirella*, while, the top section is marked by the predominance of planktonic species. These sections of the core are also classified with cluster analysis into 7 zones, in brief, the zones I to V correspond to the section with planktonic species, the zone VI to the transition segment, and the zone VII to the benthic species.

The zone VII (Fig.12) is abundant with species from the *Amphora* genus, some *Navicula* species, for example *N. cari* and *N. menisculus*, as well as *Epithemia sorex* and *Pseudostaurosira brevistriata*. In general, this zone has a much more diverse biota with diatoms from all morphologies. Nevertheless, in the most bottom part, between 320 cm and 405 cm, it is possible to find more monoraphid genera, such as *Achnanthes*, *Achnanthidium*, *Planothidium*, and *Psammothidium*. On the other hand, a high population of *Epithemia sorex* and *Cocconeis neodiminuta* found between 230 and 250 cm declines at around 300 cm, together with *Karayevia clevei*. Concurrently, there is a slight increment of the species *Amphora ovalis*, *A. pediculus* and *A. lybica* and the appearance of *Aulacoseira islandica*, and *Stephanodiscus* spp.

The transitional zone (zone VI), is characterized by a high abundance of araphid species from the genera *Pseudostaurosira*, *Staurosira*, *Staurosirella* and *Fragilaria* to a lesser degree. Some pennate benthic species start are still present, such as *Diploneis*, *Fallacia*, *Hippodonta*, *Mastogloia*, *Karayevia*, *Epithemia* and *Amphora*, together with the centric and planktonic *Lindavia rossii* diatom.

Lastly, the section with planktonic species (zones I to V – Fig. 11) has the genera *Pinnularia*, *Eunotia*, *Tabellaria* and the species *A. formosa* and *A. tenella* as the dominant. After the transition zone, the zone V displays high abundance of *A. tenella*, a reduction in abundance of *Staurosira*, *Pseudostaurosira* and *Staurosirella*, as well as a gradual increase of *Tabellaria flocculosa*. After that, the zone IV exhibits a decrease of *A. tenella*, high abundance of *A. formosa* and an increase in the genera *Eunotia* and *Frustulia rhomboides*. In the following zone III, the species *A. tenella* and *A. formosa* have a very low abundances or are absent, while *T. flocculosa*, *E. incisa* and *E. serra* are highly abundant. In the zones II and I, *A. formosa* is the dominant species, while species like *Stenopterobia curvula*, *Chamaepinnularia mediocris* and the genera *Kobayasiella*, *Melosira*, *Encyonema* and *Gomphonema* can also be found in the top.

Through the 13 samples analyzed from Storträsk, 122 species were identified to subspecies level when possible. The majority of the species are planktonic, and the most abundant group are diatoms with centric morphology (Fig. 13). The most abundant genus is *Aulacoseira*, of which *Aulacoseira tenella* is the most numerous species throughout the core (with a mean relative abundance of 56.4%). Other *Aulacoseira* species with high abundance are *A. italica* (5.5%), *A. Alpigena* (5.4%) and *A. distans* (2.5%). From the pennate diatoms, the genus with the highest relative abundance is *Tabellaria*, with *Tabellaria flocculosa* being the second most abundant species (7.5%) in the studied sediment sequence. Likewise, the genus *Eunotia* has an important relative abundance yet only *E. incisa* has an abundance higher than 2%, species such as *E. bilunaris*, *E. serra*, *E. pectinalis* or *E. meisteri* are present through most of the core too.

The species assemblages across the core show little variation, however, it is possible to separate it in two sections; from the bottom to the middle of the core. The main characteristic is an increase in *Aulacoseira tenella* from a relative abundance of around 40% to its maximum of 72% at 1.75 cm. In the second top half, the abundance of this species decreases but remains the most numerous with a prevailing abundance close to 40%. Another characteristic of the bottom segment of the core is the constant presence of *Tabellaria flocculosa* and *Brachysira bresbissonii*, while in the top the latter is absent in some samples and the first one has smaller relative abundances. On the contrary, *Eunotia incisa* and other species of the genus *Eunotia* have a higher relative abundance in the top segment of the core, as well as other species such as *Asterionella Formosa* and the genus *Stenopterobia*.

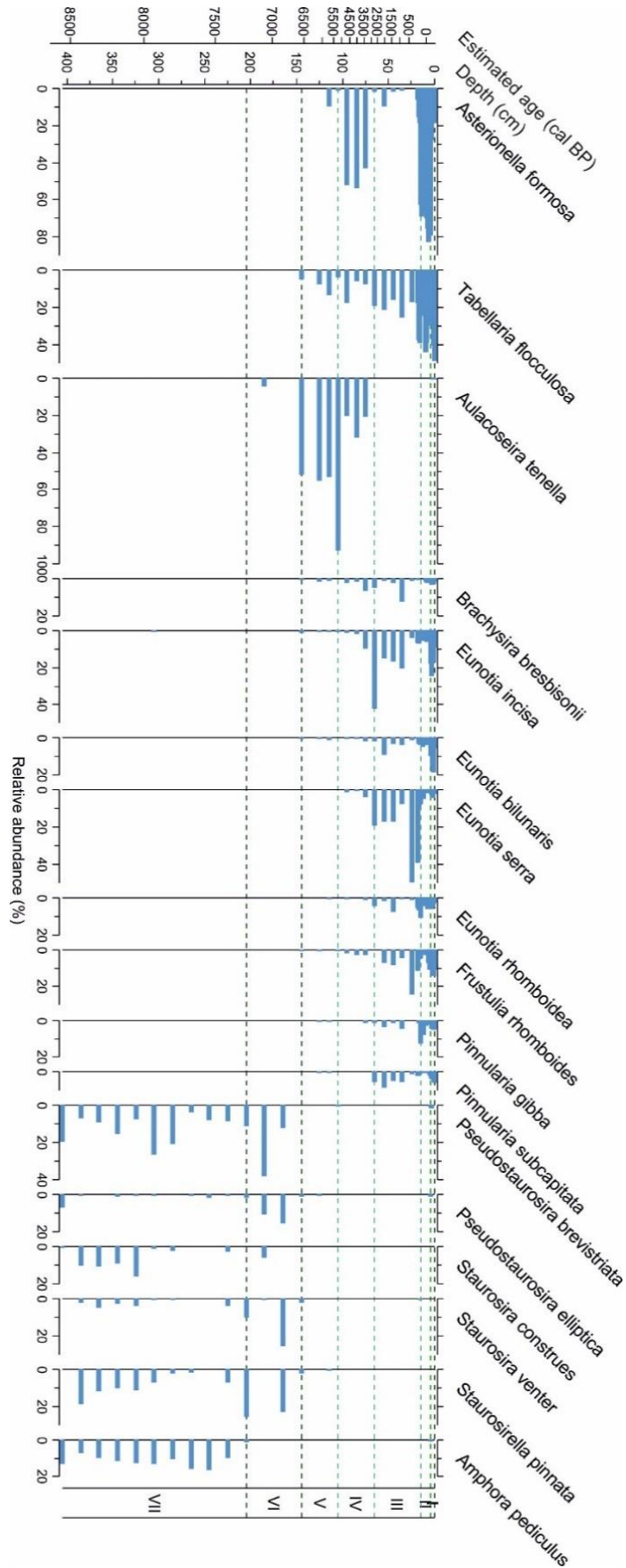


Figure 10. Relative abundance of the most common diatoms in the sequence from Hältingträsk. The cluster zones indicate three clear stages in the assemblages; benthic diatoms on the bottom, a transition zone with fragilarioid diatoms, and a final stage on the top with more planktonic taxa.



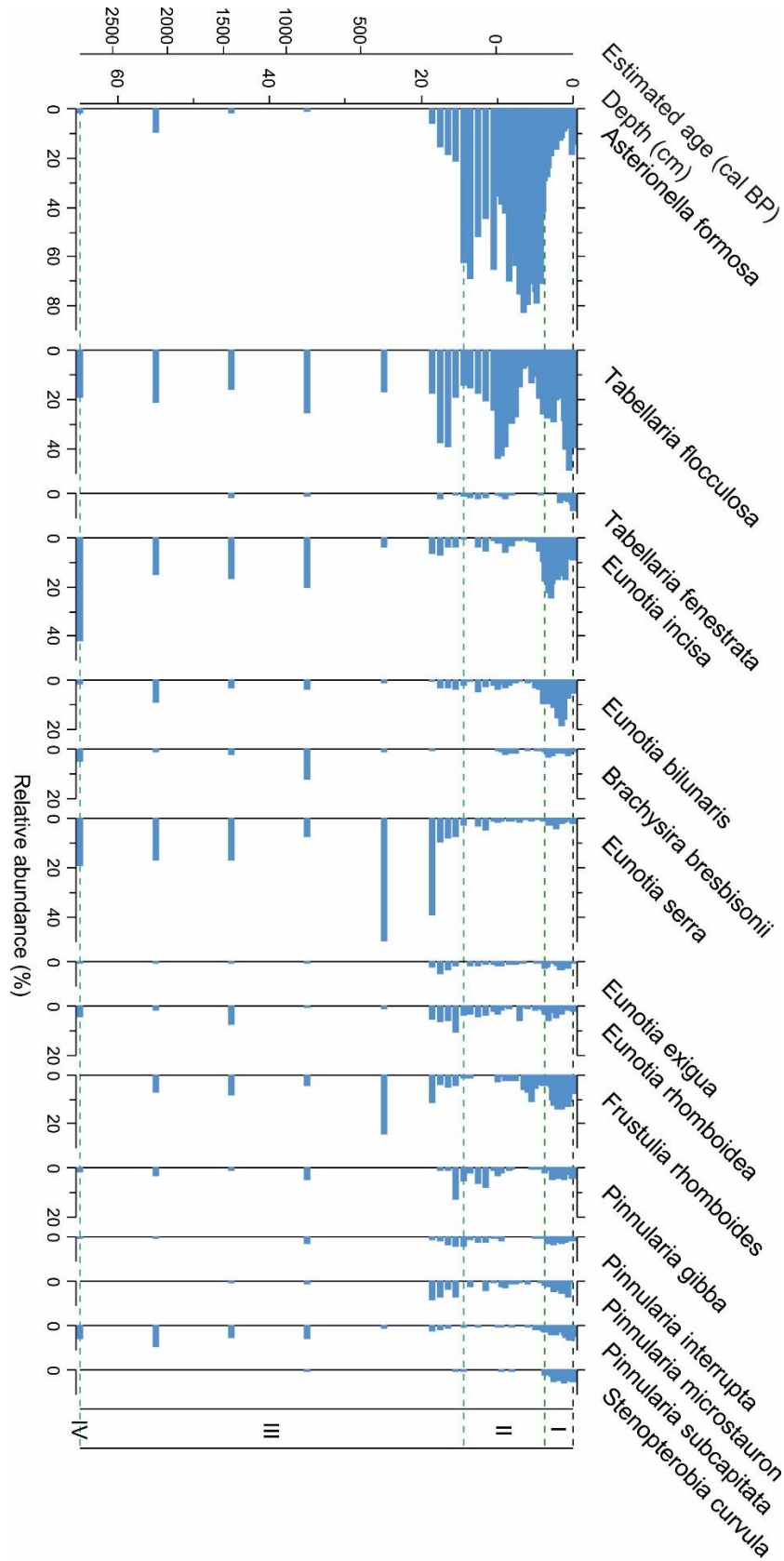


Figure 11. Relative abundance of the most common diatom species in the top 65 cm, including the 18 cm high-resolution sequence. The figure displays the diatom zones I to III.

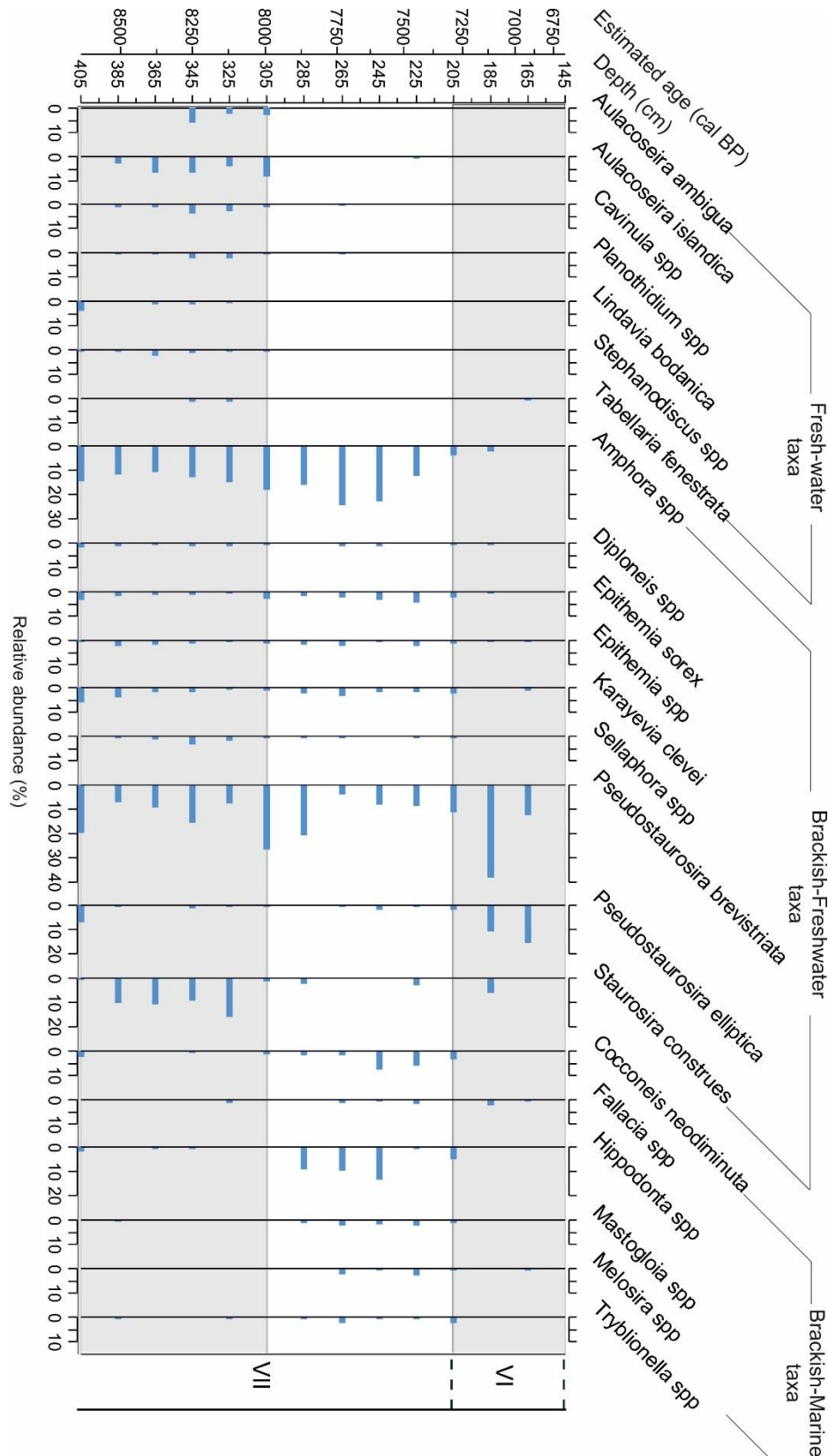


Figure 12. The zones VII and VI from the sequence have a bigger diversity than the top, the abundance of most of the species is low, therefore is not displayed in the Fig. 10. Qualitative classification of the species is used to group the species based on their salinity tolerance. Although some of these species can have broad tolerance for salinity, it was identified that the bottom 100 cm (from 405 to 305) had certain fresh-water species, that the following 100 cm (from 305 to 205) had typically marine taxa, and that some species were found throughout the entire zone VII and VI.

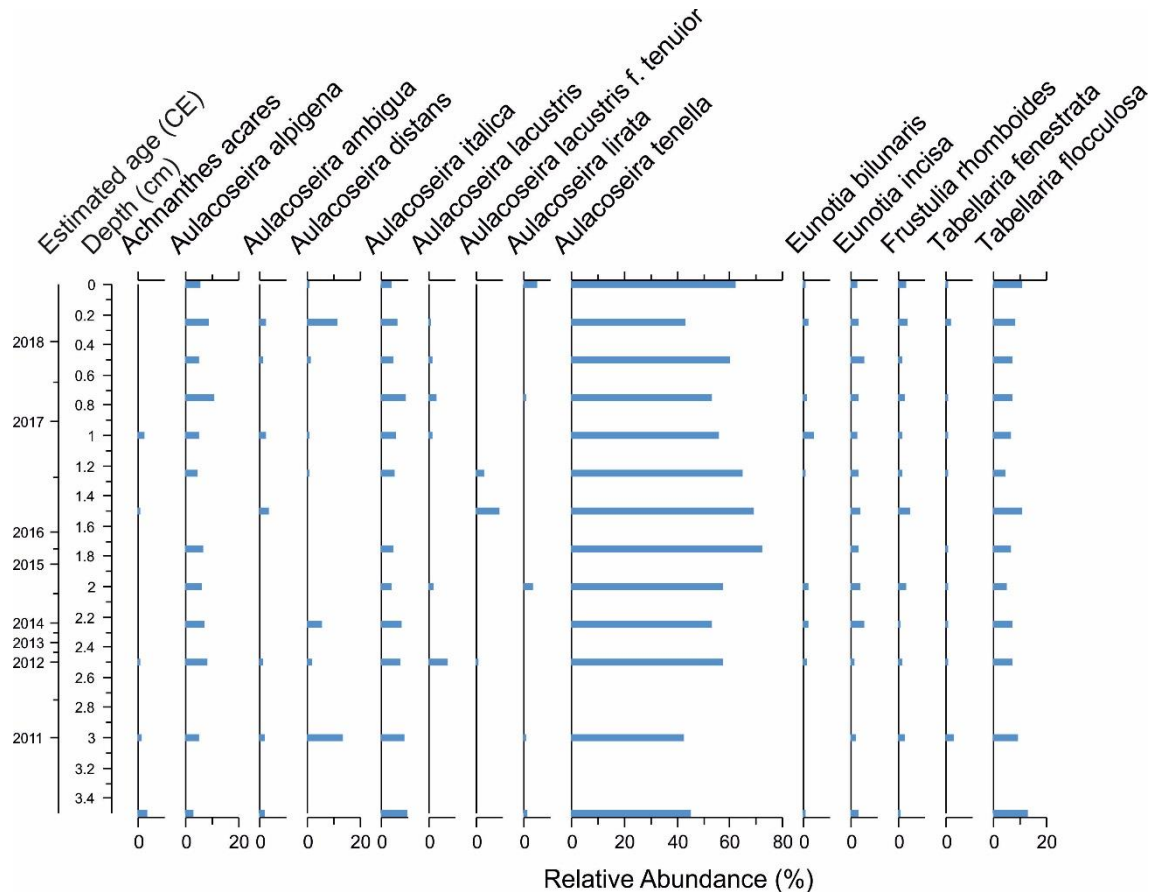


Figure 13. Relative abundances of the most common species in the core from Storträsk. This shorter sequence shows little variation and depicts the most recent changes in the lake.

## 5.4 Sediment Geochemistry

The concentrations of the 12 elements analyzed show a similar variation throughout the sediment sequence, as displayed in Figure 12. The key turning points occurs around the same depths, and these changes are classified with cluster analysis. From bottom to top, the first transition happens at the depth ~180 cm (diatom zone VII), in this zone the shifts in the geochemistry are minimum and the concentrations are comparatively low for most elements, except for Fe and Mn with higher concentrations at the bottom. The second change takes place roughly at 140 cm (diatom zone VI), in this segment the concentrations of Fe and Mn gradually decrease while Co, Cu, Cd, As, Zn and Ni concentrations increase. Between 140 cm and 110 cm (diatom V), these metals start to decrease in the same fashion as Fe and Mn, whereas V and P concentrations are higher. The section from 110 cm to 10 cm (diatom zones IV and III) show stable trends in the concentrations, excluding Ni which shows an anomalously high value at 40 cm depth. Finally, the top

most 10 cm (diatom zones II and I – Fig. 15) are characterized by two shifts defined by a peak in the concentrations of most heavy metals, in the case of Pb the maximum value of 83.92 ppm is at 4.75 cm. These changes in the top are parallel to the changes in the diatom sequence.

To assess the ICP-MS results, three replicate reference samples were measured and compared with the certified values, presented in Table 4. The elements in general have a good recovery in both reference material, except in phosphorous where the yield is very low for both reference samples, the recovery of the elements V and Cr is low too in the LKSD-4 material, but the sample WQB-1 and its duplicates show a better recovery of the certificate values. Duplicates of the 10% of the samples were measured as well, the RSD% (Relative Standard Deviation) of many of the elements has a small percentage, but the duplicate sample of the top indicates more disperse values from the mean.

Table 4. Measured concentrations of the reference material samples and their duplicates. The recovery of most of the elements was above 90%, signifying the results are trustworthy.

	LKSD-4 1	LKSD-4 2	LKSD-4 3	Average	STD	RSD%	Yield %
P	1310.83	1417.92	1529.41	0.14	0.01	6.29	21.54
V	31.07	34.06	35.54	33.56	1.86	5.53	68.48
Cr	16.49	18.11	19.06	17.88	1.06	5.94	54.19
Mn	364.73	378.79	395.94	379.82	12.77	3.36	75.96
Fe	23995.13	25106.61	26222.89	2.51	0.09	3.62	89.64
Co	9.41	9.88	10.35	9.88	0.39	3.90	89.81
Ni	30.30	31.51	32.51	31.44	0.90	2.87	101.41
Cu	29.02	28.88	30.22	29.38	0.60	2.05	94.76
Zn	205.42	184.35	193.01	194.26	8.65	4.45	100.13
As	14.99	15.99	16.95	15.98	0.80	5.01	99.87
Cd	2.25	2.05	2.20	2.17	0.08	3.91	
Pb	87.82	88.98	95.08	90.63	3.18	3.51	99.59
	WQB-1 1	WQB-1 2	WQB-1 3	Average	STD	RSD %	Yield %
P	1348.79	1261.46	1375.40	0.13	0.00	3.66	28.20
V	37.28	42.29	40.61	40.06	2.08	5.20	86.96
Cr	39.28	39.44	40.31	39.68	0.45	1.14	84.95
Mn	2016.30	1835.38	1997.64	1949.77	81.25	4.17	90.00
Fe	38773.33	37006.23	39364.75	3.84	0.10	2.61	88.86
Co	16.66	15.34	16.58	16.20	0.60	3.73	98.72
Ni	52.93	49.65	53.52	52.03	1.70	3.27	90.00
Cu	69.07	63.76	69.51	67.45	2.62	3.88	90.59
Zn	258.24	236.67	255.61	250.17	9.61	3.84	95.85
As	19.26	18.17	19.93	19.12	0.72	3.78	94.19
Cd	1.90	1.73	1.83	1.82	0.07	3.89	105.81
Pb	78.79	71.09	76.35	75.41	3.22	4.26	95.44

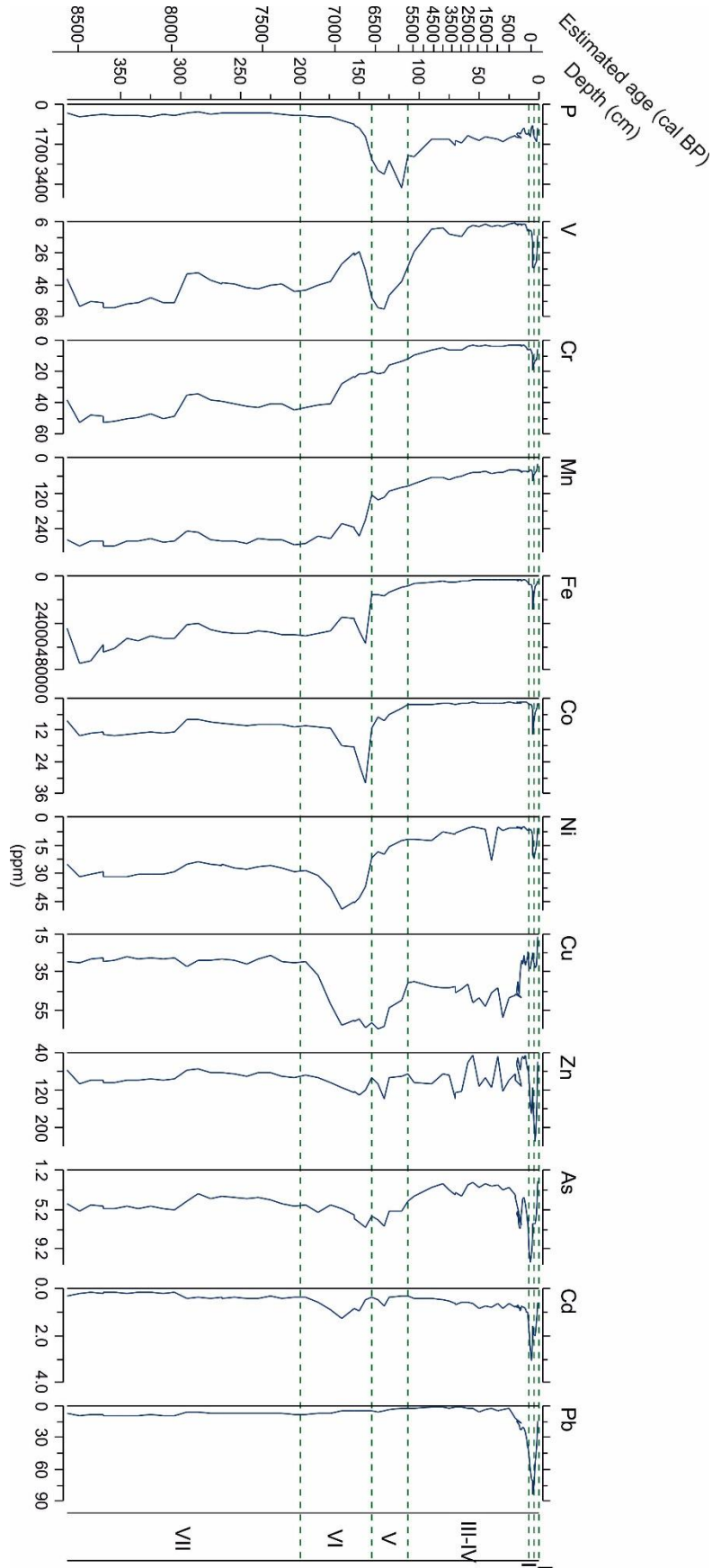


Figure 14. Heavy metals concentrations in the sediments from Hålingträsk. The cluster analysis performed on this data was in agreement with the one carried out on the diatom assemblages.

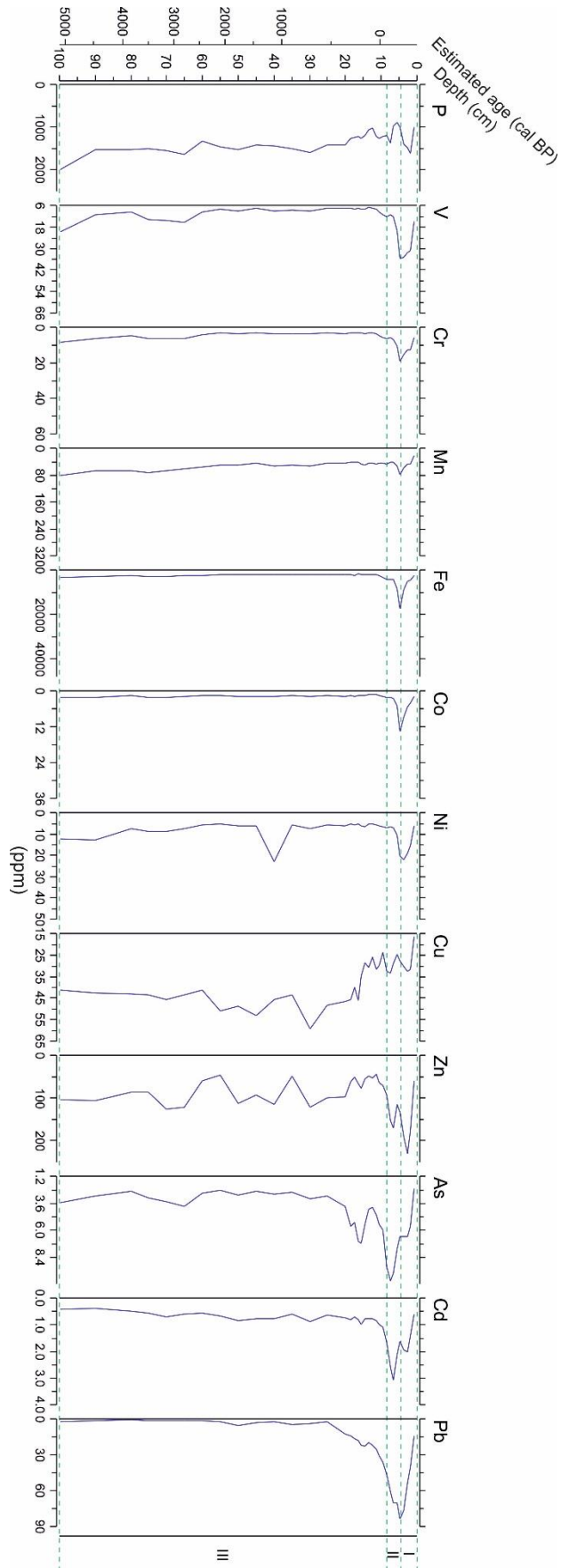


Figure 15. Trace metals concentrations of the top 100 cm, this section of the plots displays the peak of several elements.

### 5.5 Reconstructed pH

The reconstructed pH of Hålingträsk uses 119 species with more than 0.05% of relative abundance, on average, 58% of the fossil information is represented in the modern training set. However, the top 100 cm has a better representation in the training set and most of the samples have more than 70% of representativeness (Fig. 16). The bottom samples have a bigger richness of diatom species, they might be rare species uncommon in the modern training set. Nevertheless, the minDC of the majority of the samples are less than 5 percentiles away, only the samples of the bottom had a large distance to the modern analogue. In addition, R2 for Hålingträsk was 0.87, and RMSEP was 0.39, therefore the model is robust and makes a good representation of the trends in pH.

The pH in Hålingträsk shifts from alkaline to slightly acidic, following the change from a diverse benthic diatom population to a more homogenous planktonic diatom population (Fig. 17). Over the first 100 cm the values remain stable, with a pH close to 7. Then, the values start to show more variation at around 260 cm where it oscillates between 7 and 8. At the depth 180 cm, pH starts decreasing towards a slightly acidic pH. The calculated pH between 160 and 60cm fluctuates between around 5.5 and 6, while the topmost 5 cm shows a shift to a pH of ~5.5. In the top 18 cm with a high-resolution focus, the highest possible pH, is 6.5 at 14.5 cm depth, but, from the 4 cm onwards, the pH stays close to 5.5, and the lowest inferred pH would be of 5 units.

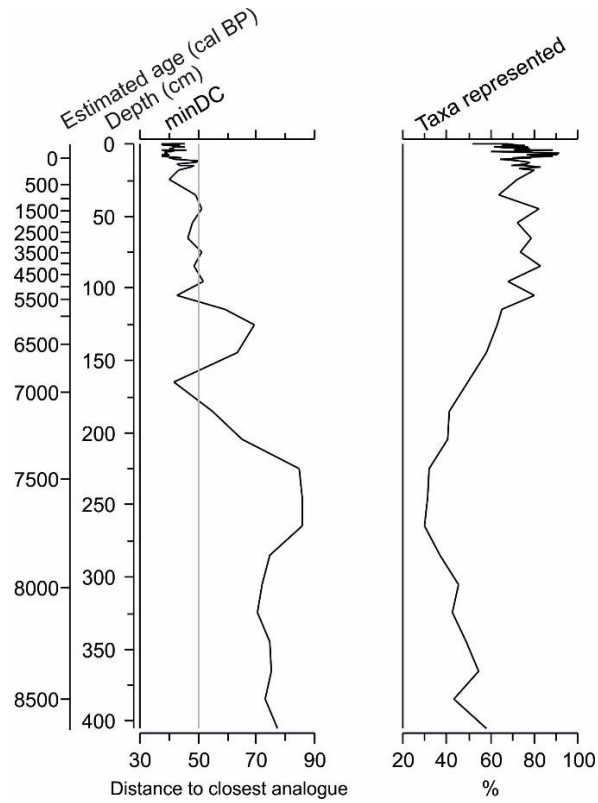


Figure 16. Distance to closest analogue in Hältingträsk with the cut-off value of 5 percentiles from the MAT. The taxa represented in the model is always above 58%, and it increases up to 70% on the top samples.

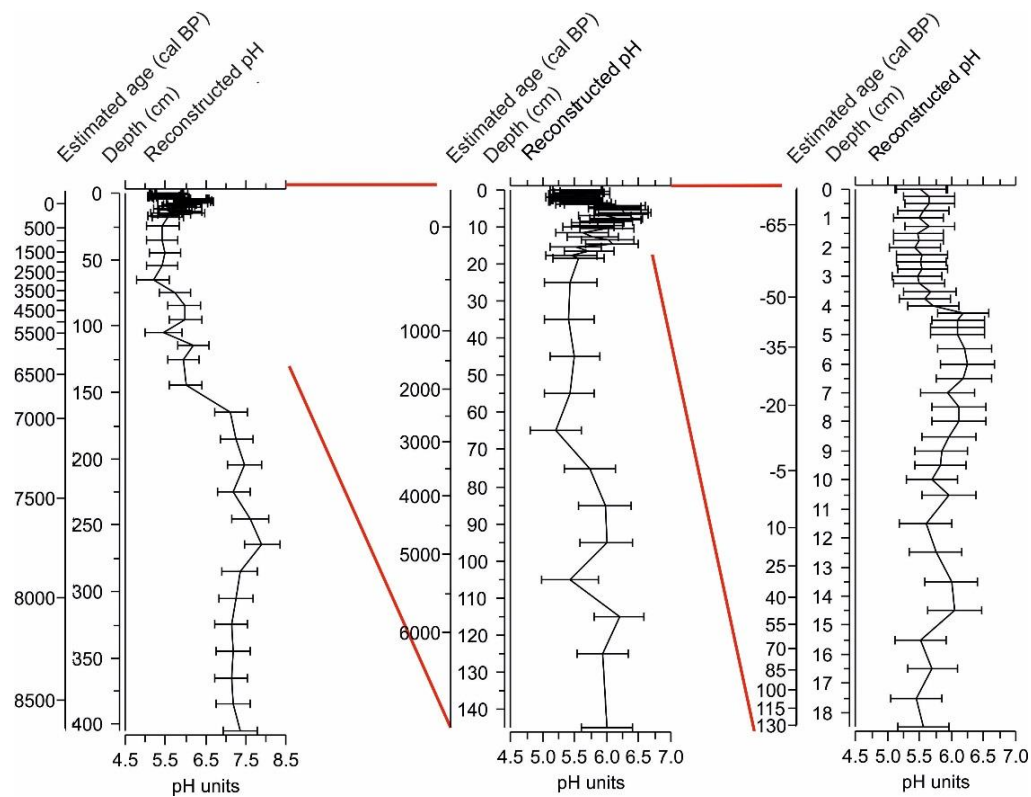


Figure 17. Reconstructed pH for different ranges of the core from Hältingträsk, the first represents the reconstructed pH for the entire sequence, the second corresponds to the zones I to V and the last one is the sequence with the highest resolution.



The reconstruction is based on the EDDI training set, which does not contain all the species present in the core from Storträsk. However, those species that are not considered for the pH reconstruction have an abundance lower than 0.025%. For the reconstructed pH, 56 species are present in the modern training set, and they are the most abundant species in the assemblage. In addition, the distance to the closest analogue from the MAT suggests that the calculated values from the reconstruction represent the trends in pH values for Storträsk, because all of them but one is less than 10 percentiles away from their modern analogue (Fig. 18). Although the MAT suggests a poor representativeness of the fossil taxa, the evaluation of the model through R<sup>2</sup> and RMSEP indicates a good model with values of 0.9 and 0.4 respectively.

For Lake Storträsk, the overall variation of the reconstructed pH is between ~4 to ~6.5. In general, the reconstruction fluctuates little and the most noticeable shifts happen in the first half. The pH reconstruction in Fig. 18 shows that at beginning, the calculated pH varies between 5 and 5.5, but in the middle of the core, from the depth 1.8 cm to 1 cm, the calculated pH is slightly more acidic and is estimated at its minimum of 4.6 at 1.5 cm. At this depth the diatom stratigraphy indicates the highest abundance of *Aulacoseira tenella* and an increase in *Tabellaria flocculosa*. The remaining reconstruction shows a stable pH close to 5.2, except for the value on the top of 5.5.

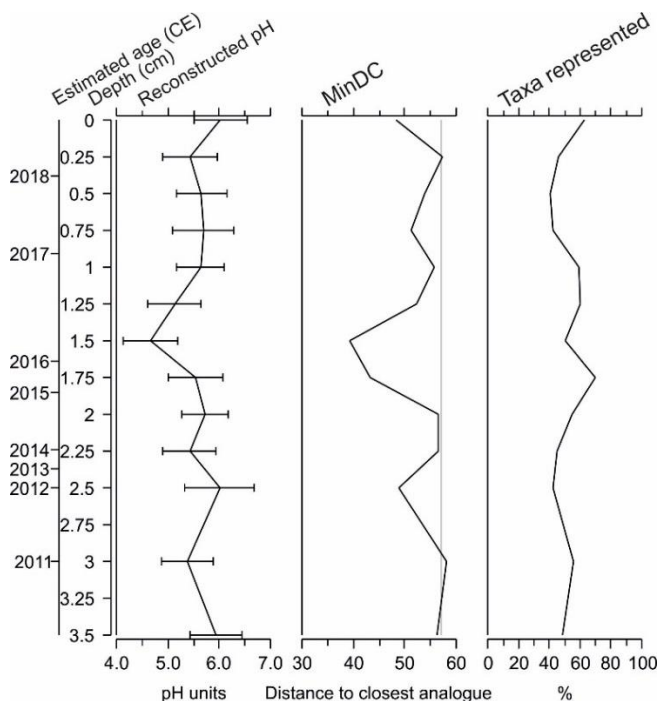


Figure 18. Reconstructed pH, and evaluation of the model through the MAT and the taxa representativeness in Storträsk.

## 5.6 Ordination and Statistics

The variation gradient from the DCA in the data from Storträsk was small in both axis 1 and 2 (1.4 SD), in consequence a linear approach could be used. On the contrary, Hältingträsk data had a larger gradient (5.5 SD) and a unimodal method was chosen for further analyses. Because of the high resolution on the top section in Hältingträsk, a separate ordination of these data was performed, the result was a smaller variation gradient (1.9 SD) that allowed to perform linear ordination. A summary of the eigenvalues is given in the following Table 5.

Table 5. Ordination eigenvalues from both DCA and PCA analyses performed in the datasets from Storträsk and Hältingträsk.

	Storträsk		Hältingträsk full core		Hältingträsk 95 cm	
	Axis 1	Axis 2	Axis 1	Axis 2	Axis 1	Axis 2
DCA	0.15	0.08	0.94	0.16	0.12	0.08
PCA	0.26	0.18			0.36	0.15

In Storträsk the variability of the diatom taxa is so small that the ordination can only explain a small percentage of the variation, so it was considered redundant. The DCA applied to the full dataset of Hältingträsk shows the three stages identified with cluster analysis along axis 1 (Fig. 19 and 20). However, only the samples from the bottom of the core show variability along the second axis and without a clear pattern related to their position in the core. In the same way, the planktonic species identified in zones IV to I are clustered together in one point along axis one while the benthic species are more disperse; the species *S. venter*, *S. construens* and *S. pinnata* are located on the top right corner while pennate species from the genera *Amphora*, *Hippodonta* or *Cocconeis* occupy a wide area from the middle to the right end of the axis 1.

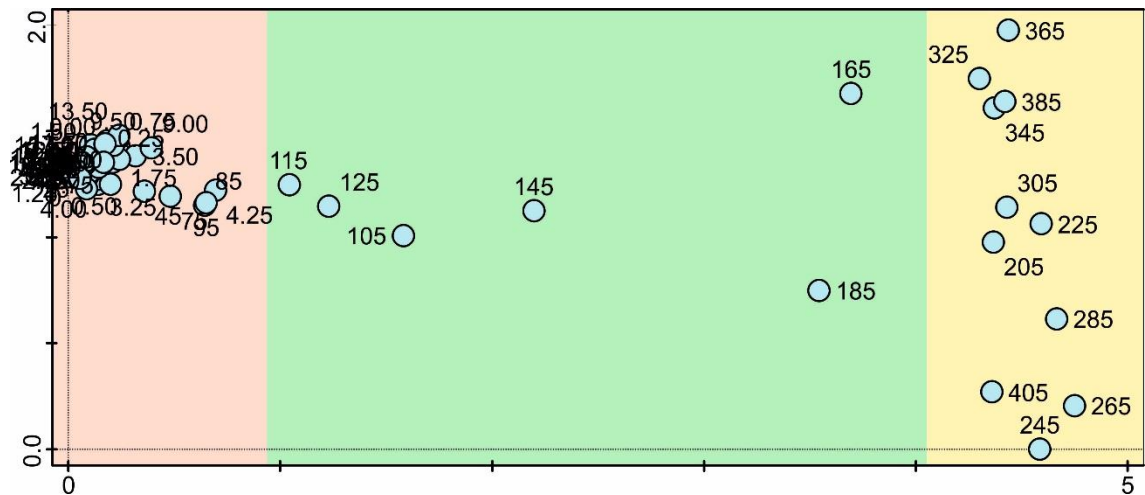


Figure 19. Plot of the DCA gradient of the samples from the entire sequence from Hålingträsk. The large gradient is in agreement with cluster analysis. The samples are grouped in three sections of the plot; the bottom samples are located at the rightmost side along the horizontal axis (yellow zone), a second transitional group is in the center (green zone), and the top samples are found in the left side of the plot (pink zone). The variation along the secondary axis is only appreciated in the bottom samples and without a specific pattern.

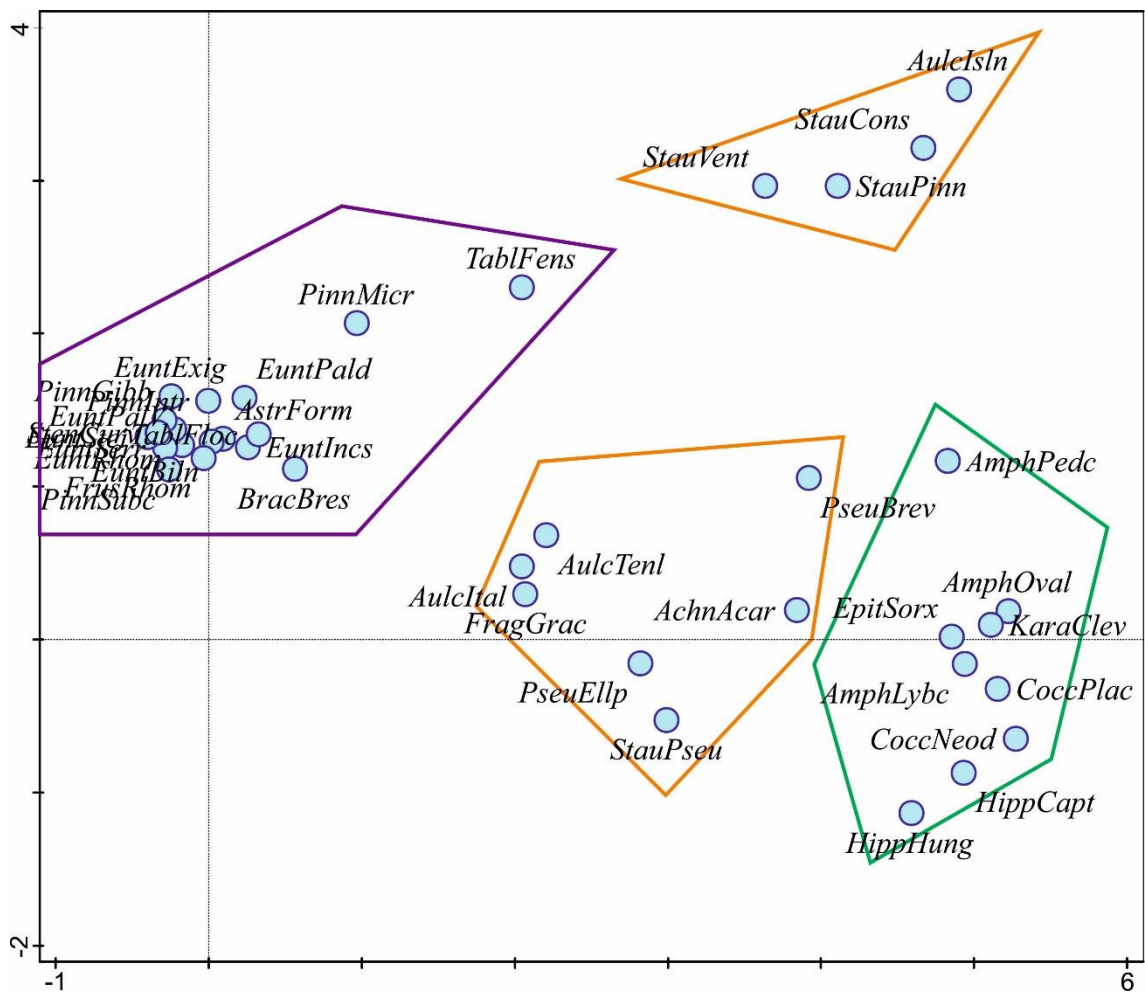


Figure 20. Plot of the DCA gradient of the diatom species from the core from Hålingträsk. Species names are abbreviated using the first four letters of genus and species name. Diatoms such as *Hippodonta* spp, *Cocconeis* spp and *Amphora* spp, are plotted in the bottom right corner (green polygon), fragilarioid species in the top center and center (orange polygons), and diatoms from acidic environments are clustered together in the left center of the plot (purple polygon).

The PCA from the zones IV to I display a better explanation of the data. The shallower samples (from 0 to 3.75 cm) are clumped together in the third quadrant, the subsequent samples (from 4 to 9 cm) are in the fourth quadrant while the rest of the samples tend to appear in positive values along the second axis (Fig. 21). The dominant species *A. formosa* stands out from the rest of the species and does not group with any other species, it is in the fourth quadrant and is the furthest away from the rest of the species. Similarly, *A. tenella* is also in the opposite side of most of the pennate species in the first quadrant, while the rest cluster in the second and third quadrant (Fig. 22).

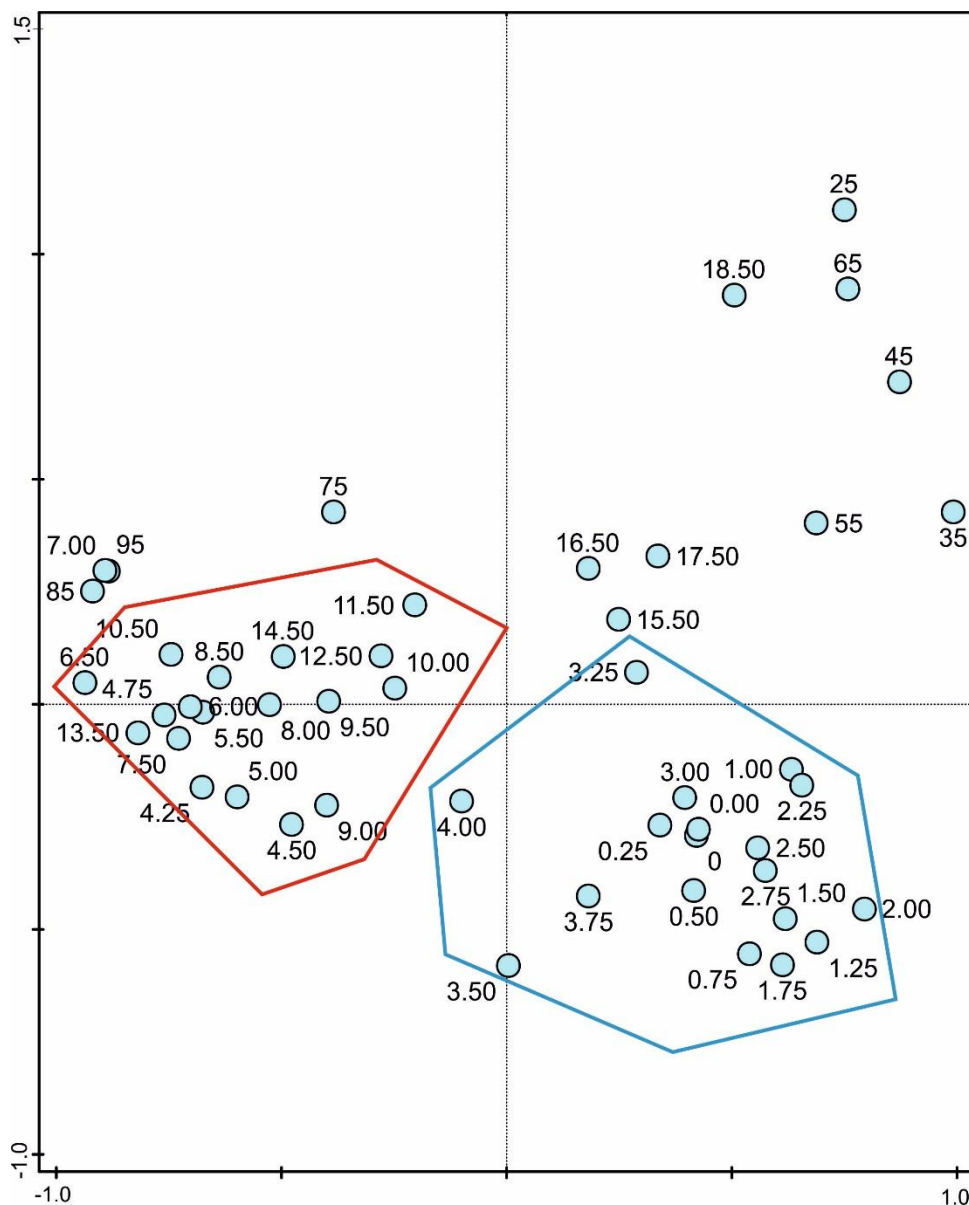


Figure 21. Plot of the PCA from the top 95 cm in Hålingträsk. The topmost 4 cm are located in the third quadrant (blue polygon), indicating their similarity among each other. The next samples, from 4.25 to 9 cm are plotted between the fourth and first quadrant (red polygon), and the rest do not have a clear distribution.

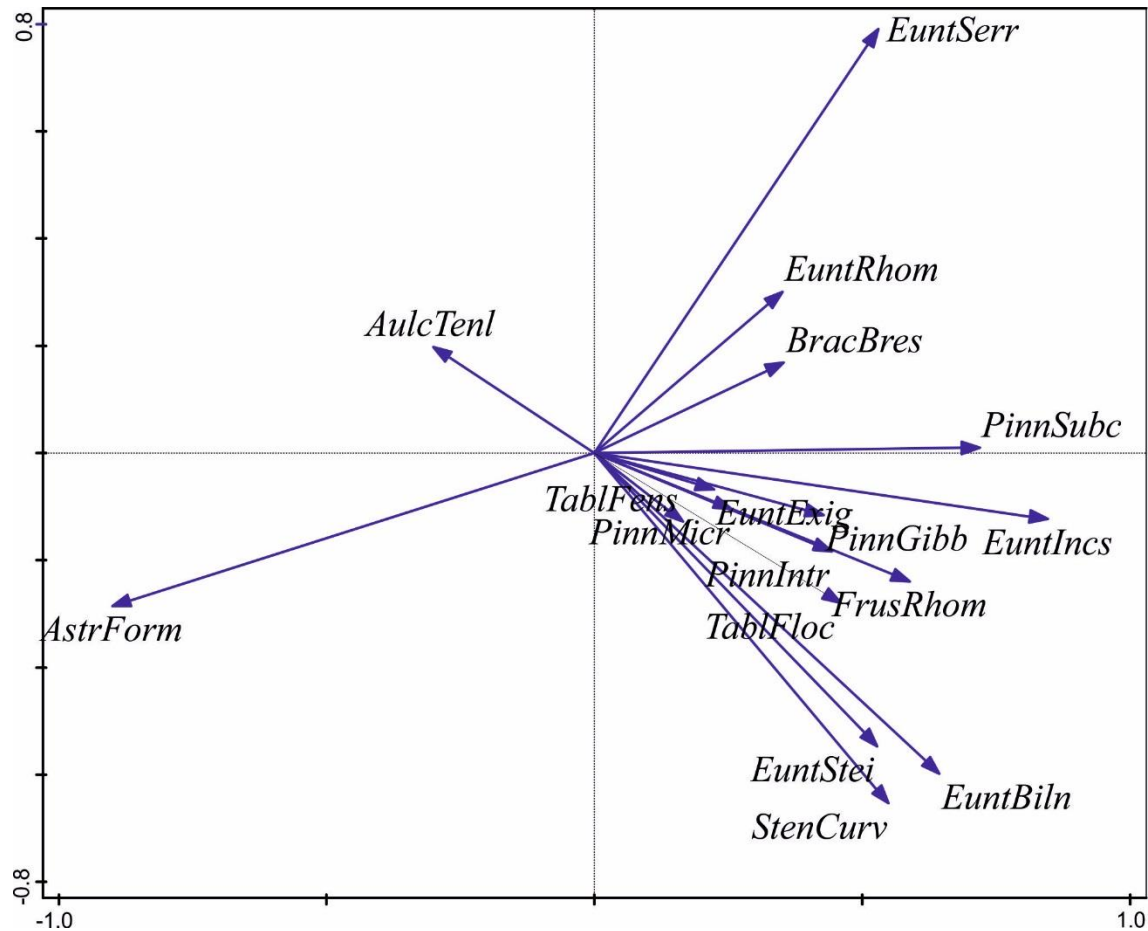


Figure 22. Plot of the PCA with the most common species from the top 95 cm from Hältingträsk. The species *A. formosa* is plotted far from the rest of the species. Although *A. tenella* is closer to the rest of the species, it is the only species in the first quadrant. The rest of the species are in the second and third quadrant.

Of the 344 species found in the sediments from Hältingträsk, most are found in the bottom, as the higher richness index shows in the Figure 20. The highest richness occurs right at the beginning of the zone VII where several benthic species start to appear, and the lowest takes place in the zone 3 when *A. formosa* has its highest abundance. Simultaneously, the evenness of the species is smaller in the bottom and tends to have less variation in the zone II, both the species richness and the evenness change in the zone V, where *A. tenella* is highly abundant. The assemblage is not as even as in the remaining zones of the core and has the biggest richness of the top segment after the transition zone VI.

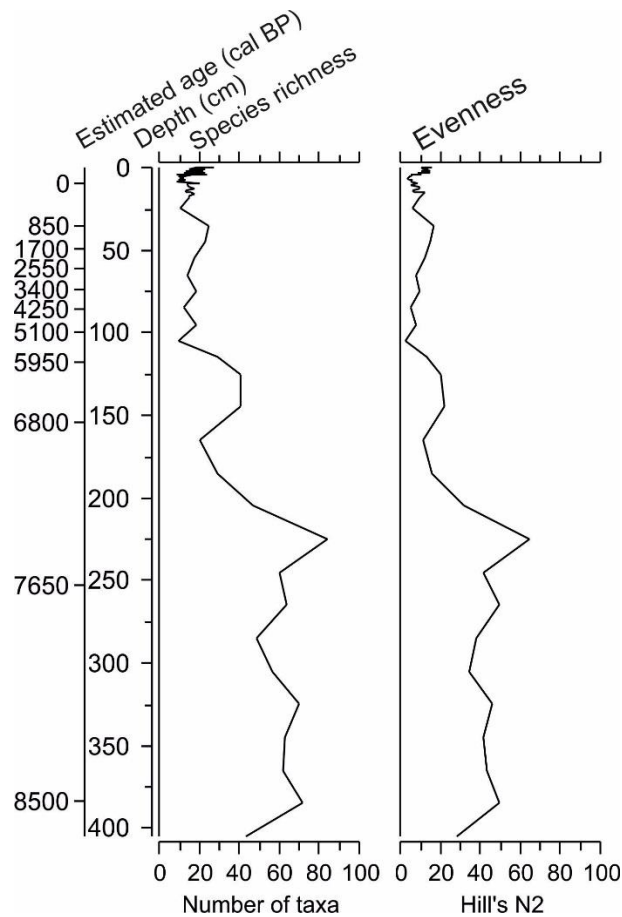


Figure 23. Species richness and evenness for Hålingträsk. The highest levels of both indices occur below 200 cm.

The total number of species identified in the sediments from Storträsk is 121, however, the majority of the samples has *A. tenella* as the dominant species (Fig. 13). The two sections are easily distinguished with the diversity index as seen in the Figure 24. In the middle (1.5 cm of depth) the richness and evenness is at its lowest, while the rest of the core shows more even diversity of species. Overall, the bottom segment has more changes in the evenness. On the contrary, the top has a more stable behavior aside from a decrease in richness in the topmost samples.

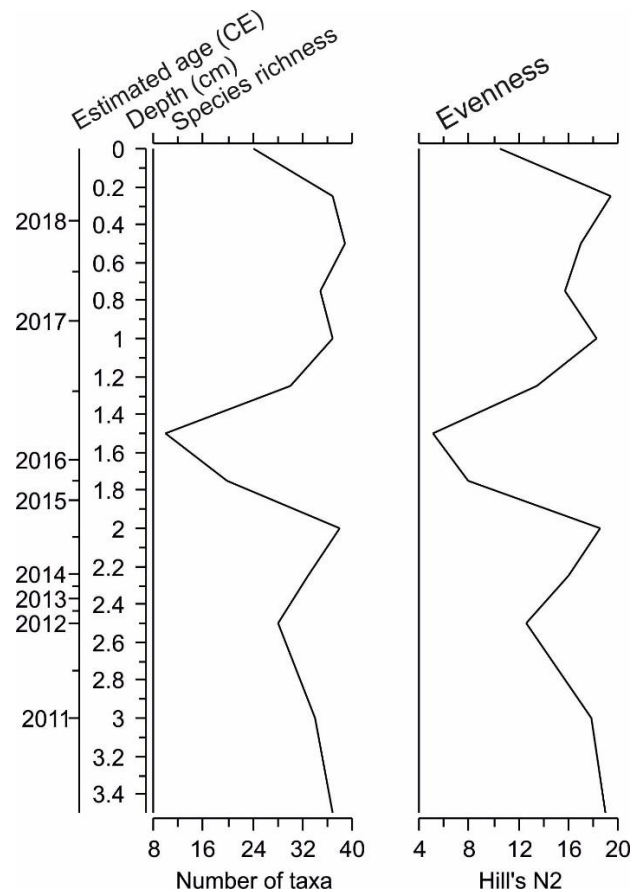


Figure 24. Species richness and evenness for lake Storträsk. The indices are heavily influenced by the high abundance of *A. tenella*.

### 5.7 Inferred Chlorophyll a

In the surface sediments sampled in summer, the photosynthetic pigments in Hältingträsk show an increasing trend through the core (Fig. 25). At the bottom of the sequence, the values of the inferred chlorophyll a are around 0.12. After a decrease between the depth 10.25 and 7 cm, the values have a steady increment until the top of the core, where the values have almost doubled.

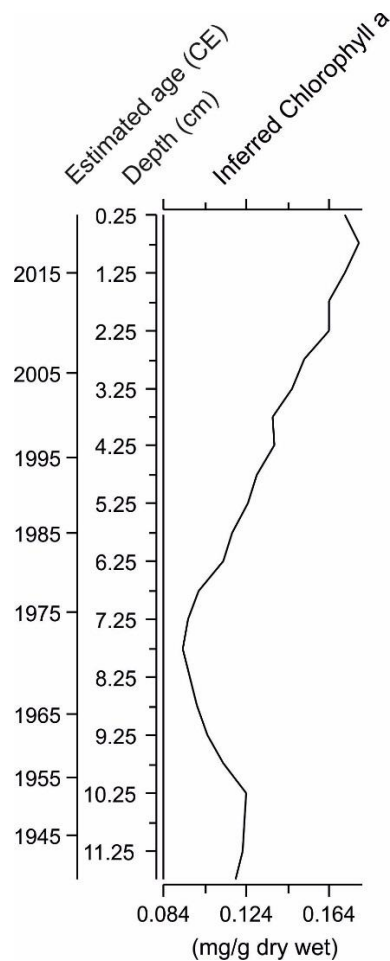


Figure 25. Inferred chlorophyll a for Hältingträsk. The general trend of the photosynthetic pigments is increasing towards the top.

The sediment sequence from the summer core in Storträsk is longer than the one from winter. However, for the top 3.5 cm the inferred chlorophyll a displays a shift from 0.06 to slightly higher values around 0.065 mg/g at the depth 2.8 cm (Fig. 26). When looking at the full sequence, the lake had low production levels of 0.050 mg/g on the bottom that gradually increased until its maximum of 0.067 mg/g at a depth of 8.5 cm. After reaching this value the chlorophyll a production started to decrease and has remained between 0.057 and 0.065.



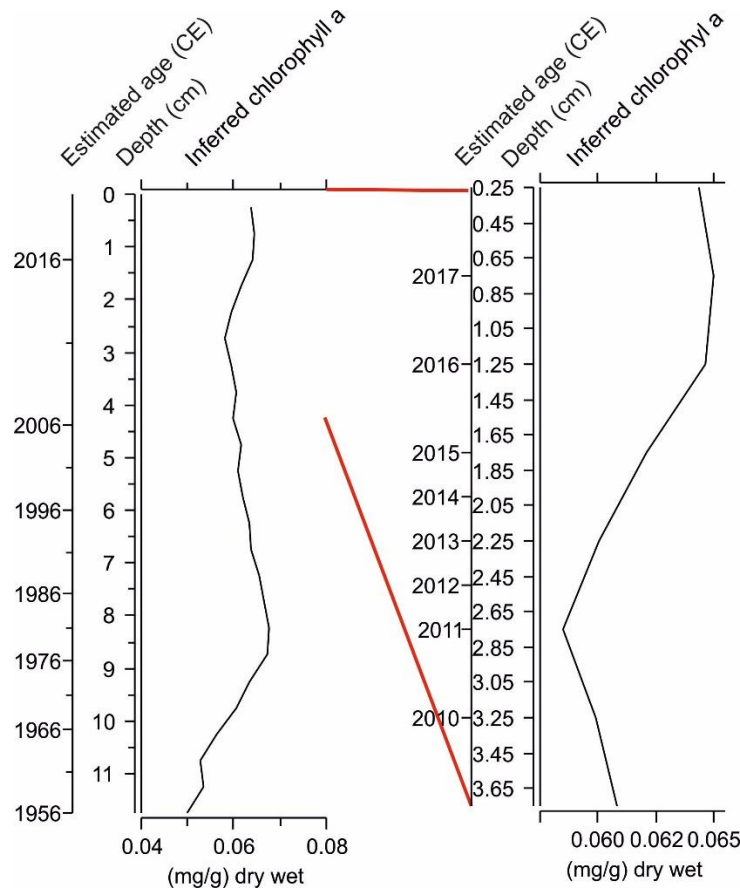


Figure 26. Inferred chlorophyll a for Storträsk. The topmost samples show a slight increase in the pigments.

## 6. DISCUSSION

### 6.1 From the Brackish Basin to the Lake Hålingträsk

The zones VII to V identified with cluster analysis from the diatom stratigraphy correspond to the period between 8600 to 6500 cal BP. This interval falls within the mid-Holocene subdivision Northgrippian, from the 8200 BP event to the 4200 BP (Cohen et al., 2018), comprising the Holocene Thermal Maximum (HTM) (Renssen et al., 2012). The geochemical record and diatom assemblages, as well as shore displacement history of the region, suggest that Hålingträsk was part of the Ancylus Lake, and later of the Litorina Sea at this time (Miettinen and Hyvärinen, 1997, Seppä et al., 2000a, Ojala et al., 2013, Kostecki, 2014). The end of the Litorina Sea stage is distinguished by a high relative abundance of fragilarioid diatoms, indicating the isolation of the lake between 7300 and 6500 cal BP (205 to 145 cm).

The Ancylus Lake covered the Baltic Basin during the period 10300 to 8200 cal BP (Björck, 1995, Miettinen, 2004, Björck et al., 2008). The water in the basin became brackish after the entrance of sea water coming from the flooded Danish Straits by the (8200 cal BP), marking the beginning of the Litorina Sea (Tikkanen and Oksanen, 2002, Miettinen, 2004). In this context, the sediments at the bottom of the studied core (405 to 190 cm) are classified as mud and mud-clay sediments (Fig. 9). They are in agreement with typical sediments in the Litorina Sea and Ancylus Lake, which are gray clays and clay-gyttja sediments with less than 20% of LOI (Miettinen and Hyvärinen, 1997, Miettinen et al., 1999). Likewise, the diatom taxa in the bottom 100 cm are typical of the Ancylus Lake (Fig.10 and 12) with species such as *Aulacoseira islandica* and *Stephanodiscus* spp, present, as well as littoral taxa that prevail in the subsequent Litorina stage, including for instance *Epithemia* spp. and *Amphora* spp. (Krishnaswamy et al., 1971, Miettinen and Hyvärinen, 1997, Miettinen et al., 1999). The concentrations of certain elements are slightly higher than in the following segment (Fig. 14). Previous studies (Manheim, 1961, Kostecki, 2014) have also found higher concentrations of certain metals in sediments from the Ancylus Lake than from the Litorina Sea, attributed to the transport of detritus and eroded material from inland during the Ancylus regression (Kostecki, 2014).

The lake has an altitude of 28 m a.s.l (National Land Survey of Finland, 2016), in this area the Litorina Shore line has been determined at its highest between 32 and 30 m a.s.l. (Seppä et al., 2000a), placing Hältingträsk below of the shore displacement curve. Supporting evidence that Hältingträsk was part of the Litorina Sea is provided by the diatom assemblages. While the cluster analysis fails to identify a change of taxa (Fig. 10), the ordination positions most of the bottom samples in the top right corner of the DCA plot (Fig. 19). Moreover, some of the common diatoms found in sediments deposited in the Mastogloia Sea and Litorina Sea are found in the core from Hältingträsk at depths between 305 and 205 cm (8000 to 7600 cal BP), including for example *Diploneis smithii*, *Hippodonta hungarica*, *Fallacia pygmaea*, *Nitzschia scalaris* and *Mastogloia baltica* (Alhonen, 1972, Miettinen et al., 1999, Seppä et al., 2000b). The introduction of *Campylodiscus clypeus* at the depth 225 cm (7450 cal BP) is also noticeable, as this diatom is a marker for the end of the Litorina Sea, commonly referred to as the Clypeus limit (Alhonen, 1972, Hyvärinen, 1980, Miettinen and Hyvärinen, 1997, Miettinen et al., 1999).

The transition from the Ancylus Lake to the Litorina Sea is also reflected in the geochemistry of the sediments. The concentrations of trace metals V, Cr, Fe, Co, Zn, As, Cd and Pb (Fig. 14), for instance, are slightly higher in the first stage than in the latter. It is common to find in the chemistry of nearshore sediments from the Baltic Sea Mn nodules with different metallic compounds (Manheim, 1961, Yli-Hemminki et al., 2016, Kuhn et al., 2017), these concretions are affected by the oxygen and the algae in the environment. As the conditions in the environment shifted from freshwater to a marine stage, it is possible the oxygenated water from the Litorina Sea facilitated the deposition of such nodules at the bottom (Kostecki, 2014).

The characteristics of the sediments and biota change between the depths 205 and 145 cm (7600 and 6500 cal BP), reflecting the isolation of the lake basin from the Litorina Sea. One distinct change is the substantial increase in organic content of the sediments (Fig. 9). This switch in the sediments has been reported in earlier isolation studies (Miettinen and Hyväerinen, 1997, Seppä and Tikkanen, 1998, Miettinen et al., 1999, Seppä et al., 2000b). Another typical sign of environmental change is the increment in the fragilarioid genera (Engstrom and Fritz, 1988, Wilson et al., 2012). Former results indicate blooms of *Fragilaria* spp. during the isolation of lakes from the Baltic Basin (Alhonen, 1972, Miettinen and Hyväerinen, 1997, Seppä and Tikkanen, 1998, Miettinen et al., 1999, Seppä et al., 2000a). In concordance with these results, the Hältingträsk sequence has the highest relative abundance of *S. pinnata* by the end of the marine stage (Fig. 10), while *P. elliptica* is more abundant towards the end of the isolation. The increase in fragilarioid diatoms correlates with peaks in some trace elements (Fe, Co, Ni, Zn, As – Fig. 14). This could be explained by the decrease in salinity and the change in vegetation of the catchment, which could have also changed the deposition conditions of the minerals (Seppä et al., 2000a, Kostecki, 2014).

## 6.2 The Ontogeny of Lake Hältingträsk

Zones V to III (145 to 14 cm) in Lake Hältingträsk record represent the lake ontogeny without human impact, while zone III (65 cm onwards) displays the state of the lake during the first human settlements and the beginning of agriculture in the region. Overall, this period registers the evolution of the lake after the isolation, the end of the

Northgrippian and the start of the Meghalayan (between 6500 cal BP and the end of the 19<sup>th</sup> century), the transition from the HTM to the neoglacial cooling, and the climatic anomalies of the late neoglacial (Korhola et al., 2000). The effects of these events on Hältingträsk can be tracked through the relationship between diatom assemblages and the environmental variables related to the evolution of the catchment, which has been widely studied before (Korhola et al., 2000, Wolfe, 2002, Smol et al., 2005, Smol and Stoermer, 2010, Rantala et al., 2015, Rantala et al., 2017).

Between 6500 and 5000 cal BP (zone V, from 145 to 105 cm), Hältingträsk was isolated from the Baltic Basin. One of the consequences was the change from sediments with high mineral content, to sediments enriched in organic compounds (Fig. 9). Besides, the climate was warmer and drier in southern Finland than it is today under the HTM (Johnsen et al., 2001, Tiljander et al., 2003, Seppä et al., 2009, Luoto et al., 2010). The associated shorter ice cover period and longer growing season likely resulted in higher productivity and, as a consequence the sediments report higher organic carbon content with LOI values between 40 and 65%. Since the lake isolation, the concentrations in trace metals started gradually decreasing because of the absence of manganese nodules from the Litorina Sea (Fig. 14). However, the new conditions continued to facilitate the mechanisms of removal and stagnation of minerals that allowed the deposition of metals in the sediments of the lake (Manheim, 1961, Piper, 1994, Sternbeck et al., 2000). Overall, these results indicate changes in the physicochemical conditions of the lake.

After the isolation (~5000 cal BP – 105 cm), there is a clear shift in pH towards a more acidic environment in comparison with the neutral-alkaline environment of the brackish basin (Fig. 17). The decline in the reconstructed pH is the outcome of the fading signal of the typical benthic Litorina Sea diatoms, as well as the gradual appearance of the fresh-water diatoms from the genera *Eunotia*, *Brachysira*, *Pinnularia* and especially *Aulacoseira* (Fig. 10). Still, in the early times of the lake formation there is still some imprint of the marine littoral taxa with diatoms such as *Nitzschia* spp, fragilarioids and *Caloneis* spp (Miettinen and Hyvärinen, 1997, Seppä et al., 2000a). The predominant *A. tenella* is a common species in waters with neutral to slightly acidic pH (Siver and Kling, 1997) from oligo-mesotrophic lakes, and can represent more than 40% of the taxa in oligotrophic lakes (Bicudo et al., 2016), as it is the case of the lake throughout the zone V. The high abundance of *A. tenella* and other planktonic *Aulacoseira* species could be

related not only to the nutrient input from the newly exposed soils, but to the mixing and high turbulence in the lake due to the short ice cover and the reduced vegetation in the developing catchment (Weyhenmeyer, 2007). Additionally, the water level of Hältingträsk was probably higher than at present, according to lake level reconstructions from southern Finland (Sarmaja-Korjonen, 2001, Luoto et al., 2010). The typical lake succession suggest deeper oligotrophic lakes after their formation (Engstrom and Fritz, 1988, Engstrom et al., 2000), and this is represented in the records from Hältingträsk.

Between 5500 and 3250 cal BP (zone IV), the lake is in a stage of transition, depicting the end of the HTM. Earlier research suggest that progressive cooling began in northeastern Europe around 3500 cal BP (Salonen et al., 2011), that by 3000 cal BP temperatures were already colder in southern Finland (Mäkilä, 1997, Tiljander et al., 2003) and that the forest changed from a mixed coniferous forest to a boreal forest (Seppä et al., 2009). Such changes to a moister and cooler environment, and the parallel change in vegetation have been reported to correlate with the formation of peatlands and the decrease of pH (Fig.17) (Korhola, 1995, Seppä and Weckström, 1999).

After the early phase of lake ontogeny there is a period (5800 to 3600 cal BP) where the reconstructed pH has a stable value around ~5.8. However, towards the end of zone IV there is a slight decline to ~5.2 (Fig. 17), paired with a gradual increase in the organic carbon deposited. This shift is a result of the emergence of freshwater diatom community and the end of the marine influence, which is confirmed by the PCA analysis (Fig. 21 and 22). The most abundant species through zone IV is *A. formosa* (Fig. 10 - 11), usually associated with increasing nutrients input (Wolin and Stoermer, 2005, Neil and Gajewski, 2017). Nevertheless, the prevalence and still high abundance of *A. tenella* suggests the lake is still oligo-mesotrophic. Additionally, there is a small increase in *T. flocculosa* and *Eunotia* spp, both frequently found in slightly acidic waters and the latter is also associated with bogs (Lange-Bertalot, 2001, Bouchard et al., 2017).

Based on archeological findings near the shore of Lake Hältingträsk and palynological evidence, the first human settlements began in the Late Bronze Age around 3000 cal BP (1000-900 cal BP) (Sarmaja-Korjonen, 1992). Accordingly, the beginning of zone III (~2700 cal BP) registers the early human activity and associated changes in Hältingträsk. This period of time (2700 cal BP until the end of the 19<sup>th</sup> century ) also encompasses two climate anomalies; the Medieval Warm Period (900-1300 AD) and the Little Ice Age

(1550-1900 AD) (Tiljander et al., 2003, Luoto et al., 2010). After the transition in zone IV, the zone III shows less dynamic fluctuation in the studied environmental variables and could be considered as the conditions prior industrialization and pollutant atmospheric input.

Following the lowering of pH at the end of zone IV, the pH remained acidic at around 5.4 during much of zone III. At the same time, the diatom evidence allude to the complete establishment of the bog surrounding the Lake Hältingträsk, for instance the proliferation of *Eunotia* spp, *Pinnularia* spp and *T. flocculosa*. The influence of the shoreline vegetation over the diatoms in the lake has been documented before (Bouchard et al., 2017), and diatoms from the genera *Eunotia* and *Pinnularia*, found in zone III, are common in *Sphagnum* moss and environments with low pH (Lange-Bertalot, 2001, Liu et al., 2011, Bouchard et al., 2017). The simultaneous disappearance of *A. tenella* from the record, further confirms the hypothesis of the formation of the peatbog, as the recently established *Sphagnum* moss and the change in forest vegetation lowered the pH (Fig. 17). In addition, the LOI reports high values between 75 and 85% (Fig. 9), signifying a high organic content deposition, this increase in LOI associated with peat formation in Finland has been noted before (Timoney et al., 1993, Korhola, 1995).

The geochemistry of the zone III also shows a small increment in the concentration of Cu, Zn and a peak in Ni (Fig. 14 - 15), the same momentary increase in Cu and Zn have been reported in Storträsk to a lesser degree (Rantala, 2013). Agriculture became present around 2400 cal BP and, at this time a phase of forest clearance has been reported by previous charcoal studies (Sarmaja-Korjonen, 1992). Preceding research on forest fires has not found a correlation between fires and changes in the elements deposited in lake sediments (MacDonald et al., 1991), but the Cu, Ni and Zn have been linked to erosion of soils and wastewater discharge (Zhang et al., 2015). Soil erosion could cause an increase in nutrients, which would correlate with the manifestation of *A. formosa* (Neil and Gajewski, 2017). Further evidence on the effects of the agriculture has been registered in other lakes in Sipoo (Sarmaja-Korjonen, 1992, Luoto, 2009, Rantala, 2013). For instance, an increase of LOI in Hampträsk is attributed to nutrient input from the crops (Luoto, 2009), also experienced in Hältingträsk and Storträsk (Rantala, 2013). It is possible that the LIA is registered in the record towards the end of the zone III (~1900 AD), where the diatom *A. formosa* decreases (Fig. 10 - 11), pointing a decline in the

nutrient input and catchment erosion due to unsuccessful crops suffered in southern Finland because of the low temperatures during this period (Luoto, 2009).

### 6.3 Recent Times and Consequences of Industrialization

The top centimeters of the sequence from Hältingträsk have the highest concentrations in heavy metals, the biggest shift in the diatom community and changes in lake productivity. Altogether, the last two zones depict evidence of the industrialization in Europe and urbanization near Sipoo with the rise of Pb and oscillation of some diatom species, as well as the anthropogenic warming in the increasing photosynthetic pigments and *A.formosa*. Zones II and I are proxy for the population growth that followed the new industrial society of the mid-18<sup>th</sup> to early 19<sup>th</sup> century. The accelerated change in the sediments reveal an equally fast pace in the events during this period of time.

The trend of increasing heavy metal deposition in lakes at the beginning of the late 1800s has been reported in studies all over Europe and North America for more than 30 years (Tolonen and Jaakkola, 1983, Davis et al., 1985, Verta et al., 1989, Battarbee, 1990, Virkanen et al., 1997). Hältingträsk is not an exception, cluster analysis classified the top 8.5 cm of the heavy metals in two zones; the peak and the decrease (Fig. 15). However, elements such as Pb, Cd, As, Zn and V start their increment already from the depth of 15 cm (1890 AD). The deposition mechanism of Pb, Zn and Cd has been proven to be related to long distance transport (Verta et al., 1989). Likewise, the enlargement of Pb, Zn and Cu in lakes is connected to the European industrialization that started after 1840 and to atmospheric pollution (Hakala and Salonen, 2004). Hence, the increment of these trace metals can be explained by atmospheric input.

In contrast to Pb, Zn and Cd, the origin behind the deposition of Cu, Ni and V are attributed to local sources (Verta et al., 1989). Ni and V began their increment at a depth of ~10 cm when the rest of the metals were at their maximum. At this point, Cu oscillates after the decline at the end of the zone III associated to the relapse in agriculture (Fig. 15). Synchronously, in the border between zones III and II, there is an increase of the species *A. formosa* and the emerge of the genus *Melosira* in some samples, both species are related to higher nutrient conditions (Kilham, 1990, Wolin and Stoermer, 2005, Neil and Gajewski, 2017). The increment of these elements and diatoms, between 14 and 10

cm (1950 and 1930 AD), could be a sign of a resurgence and intensification of agricultural practices near Hältingträsk.

Characteristics of the Lake Hältingträsk, such as the granitic bedrock, a small catchment, the peat bog vegetation soils surrounding the lake, and the atmospheric loading of acidifying substances and trace metals, would lead to think the water would have a lower pH (Battarbee, 1990, Korhola and Tikkanen, 1991, Tarvainen et al., 1997). While there are signs of acidification, such as the maximum of the acidophilus species *E. serra* and *F. rhomboides* in the limit between zones III and II (Fig. 10) (Camburn and Charles, 2000, Shinneman et al., 2016), it seems the lake-water was undergoing a process of alkalization (Fig. 17). Analogous episodes of increasing pH have been described in North America (Norton et al., 1988) and in naturally acidic lakes from southern Sweden (Renberg et al., 1993, Bindler et al., 2002), attributed to sulfur deposition and soil erosion. This statement is backed by the hypothesis that the fires for the slash-and-burn husbandry practices in Sweden caused the destruction of biomass and humus in soil layers (Renberg et al., 1993), and thus facilitated an increase in the pH. Bindler (2002), however, argues that the alkalization in Swedish lakes was induced by sulfur deposition through weak acid rain. Moreover, studies from Finnish lakes have related rising lake-water pH and rainwater in acid lakes (Korhola and Tikkanen, 1991). For these reasons, it is feasible that a combination of factors have caused a cultural alkalization of Hältingträsk; the loss of acid sulfate soils, common in deposits from the Litorina Sea (Tarvainen et al., 1997), by agriculture, combined with the sulfur deposition from atmospheric input. Nevertheless, further research on the cycle of sulfur on the lake is needed to assure this supposition.

The increment of organic content in the sediment after the LIA and mid-18<sup>th</sup> century further confirms the effect of increasing cultivated land. The LOI remained higher than 60% through the Meghalayan with some minor oscillations (Fig. 9). Nonetheless, looking at the top 20 cm, there is a peak around 10 cm, simultaneous to the maximum of the heavy metals (Fig. 9 and 15). In similar fashion, the LOI in nearby lakes Hämträsk (Luoto, 2009) and Storträsk (Rantala, 2013) also showed an increase in organic matter related to the nutrient increment from the agriculture. In Hämträsk, the slash-and-burn cultivation resulted in an increment in the total phosphorus and therefore, the lake became eutrophic (Luoto, 2009). By the end of the zone III (end of 19<sup>th</sup> century), *A. formosa* and pH reached its maximum too (Fig. 11 and 17). These increments could be attributed to agricultural



activities and its following enrichment in N, or warming climate like the one experienced nowadays. Recent studies argues the introduction and rise of *A. formosa* are related to the current warmer climate (Sivarajah et al., 2017). Additionally, *A. formosa* has been linked with cultural eutrophication in lakes too (Stager, 2018). Finally, another proof of the rise in the trophic level is the growing trend in the reconstructed chlorophyll a (Fig. 25). Overall, the Lake Hältingträsk features signs of eutrophication probably due to intensification in agricultural practices since the early 19<sup>th</sup> century. And, as similar cases from the area (Luoto, 2009, Rantala, 2013) and North America (Stager, 2018), it does not shows signs of a full recovery, for eutrophication can continue long even after reduced nutrient input (Stager, 2018).

The last segment of the core displays high variability. For instance, *A. formosa* started decreasing at 4 cm of depth, likewise did the heavy metals concentrations and the organic content in the sediments (Fig. 9, 11 and 15). On the other hand, the reconstructed pH declined due to the relative increment of diatoms from the genera *Eunotia*, *Pinnularia*, *Frustulia*, *Brachysira* and the species *T. flocculosa*, all frequent in acidic waters. Meanwhile, the highway Valtatie 7, connecting Helsinki with Kotka and passing Sipoo and Porvoo was constructed around 1930. Also, it is presumed that the ditching in Hältingträsk was introduced around 1950s. Meanwhile, the agriculture has not stopped. This last segment reflects the most recent times in Hältingträsk, as well as the new alterations to the catchment and close by source of atmospheric input.

The decrease in the concentrations of trace metals in lake sediments has been attributed to the reduction of Pb in gasoline (Verta et al., 1989, Battarbee, 1990). Therefore, regardless of the new highway close to Sipoo, the concentrations of heavy metals in the sediments has decreased and, in most cases, reached the levels of the early 1800s. In addition, the drop of trace metal levels could also be attributed to modernization of agriculture. After 1995 the Finnish Agri-Environmental Programme (FAEP) started and decreasing loading of nutrients to water bodies became a priority (Rekolainen et al., 1999, Vuorenmaa et al., 2002). While the decline of inorganic fertilizers happened in the early 1990s (Rekolainen et al., 1999, Åström and Spiro, 2000, Granlund et al., 2005), metals such as Mn, Fe, Co and Pb show decreasing trends after this decade in the record (Fig. 15). The reduction of atmospheric pollutants and leaking of toxic metals might have mitigated the alkalization processes throughout zone II, as the zone I documents a

decrease in ~0.7 pH units (Fig. 17), this state is comparable to the acidic pH prior the intensification of the human impact.

There are research antecedents of the connection between ditches and changes in the ecological status of lakes in boreal regions (Turkia et al., 1998, Marzecová et al., 2017, Tammelin et al., 2019). Ditching and fertilization of Finnish forests became a common practice at the beginning of the 20<sup>th</sup> century to improve agriculture and forestry (Soininen, 1974, Turkia et al., 1998, Tammelin et al., 2019). Additionally, it has been register that the drainage of peatlands increases the organic content and the acid runoff in lakes (Tarvainen et al., 1997, Turkia et al., 1998, Tammelin et al., 2019). Since the year 1990 (zone I), the LOI percentage is almost 10% less than in zones III and II (Fig. 9), similar drops in organic matter of sediments have been reported from lakes undergoing shore and catchment erosion (Luoto, 2009, Stager, 2018). However, it is not possible to conclude if the decline in LOI is related to soil erosion, because the organic content is still high, and the decrease could be a transient trend. All in all, another possible explanation behind the acidification of Hältingträsk in the recent years is the ditching.

The modernization of agriculture meant not only a reduction in fertilizers and nutrient input, the landscape has been modified too. The open ditches from early 1950s changed to hidden ditches during the 1970s to 1980s (Hietala-Koivu, 2002). The cultural eutrophication, consequence of agriculture nearby at the beginning of the 19<sup>th</sup> century still prevails. But at the same time, *A. formosa* decreased after the decade of 1990 (4.75 cm Fig. 11). It is possible that the reduction of nutrients discharge was a consequence of the effect of erosion control through the disappearance of open ditches (Rankinen et al., 2015). Supplementary factors are the minimization of fertilizers (Rekolainen et al., 1999, Vuorenmaa et al., 2002, Granlund et al., 2005) and the impoverishment of soils in past decades, due to nutrient losses (Vuorenmaa et al., 2002, Taka et al., 2016). Although *A. formosa* is on decline, it is still present and abundant, further, the levels of chlorophyll a are rising (Fig. 11 and 25). This evidence implies the productivity of Hältingträsk is still high and typical of eutrophied lakes. The slow response of lakes to agricultural modernization has been argued before in previous research, and a restoration of nutrient levels is expected to take years (Granlund et al., 2005, Ekholm et al., 2007, Stager, 2018)

Despite the fact of an apparent restoration in the water pH, the algae community is different to the one before the acidification and the alkalization. While the genera *Eunotia*,

*Pinnularia*, *Brachysira* and species *T. flocculosa*, common in zone III, are still present, the top samples also register new species such as *Chamaepinnularia mediocris*, *Kobayasiella subtilissima* and *Ecyonema* spp. Conjointly, the relative abundance of *A. formosa* is still higher than in zone III and the genus *Pseudostaurosira* has reappeared (Fig. 11). Sivarajah (2017) has ascribed this kind of taxonomic shift after pH recovery to climate warming. Although a rise in the temperature does not have a direct impact in the diatom assemblage, the climate forcing has an effect on different aspects of the lake properties; for instance shorter ice coverage, longer growing season and imbalance of the mixing and stratification of the lake (Smol et al., 2005, Rühland et al., 2015, Sivarajah et al., 2016, Sivarajah et al., 2017). In Finland, the ice cover season has become shorter throughout the last decades of the 19<sup>th</sup> century (Korhonen, 2006), entailing warmer winters and longer bloom seasons. In this regard, recent models link the increment of Chlorophyll a not only to nutrients and P from runoff, but also to warmer water temperatures (Rankinen et al., 2019). The growing concentrations of Chlorophyll a in the top samples could be influenced by the cultural eutrophication and climate warming (Fig. 25). It is likely that the shift in the diatom community is a sign of the current rising temperatures from climate change, as well as a sign of rehabilitation of the acidic pH of the lake.

The Lake Hältingträsk does not count with a fish population at the moment nor natural nor from fish introduction (Pellikka, 2018). Fish are sensitive to environmental changes and abiotic factors (Rosseland and Henriksen, 1990, Fausch et al., 1990, Tammi et al., 2003, Charifson et al., 2015). To this regard, the reconstructed pH from the topmost samples (top 4 cm) corresponds to the modern measured pH in the lake. While the reconstruction points to an average pH of 5.5, the records indicate values between 4 and 5.5 (Pellikka, 2018). A diminution in the pH has been an explanation for the loss of fish in Nordic countries (Rosseland and Henriksen, 1990, Tammi et al., 2003). The literature suggests fish are susceptible to low pH and modifications to their physical habitat. Also, that loss of fish population can happen in less than 10 years, especially under fast changes of water chemistry (Davis et al., 1985, Rosseland and Henriksen, 1990). Hence, the recently acidification in Hältingträsk of almost 1 pH unit can account for the absence of fish in the lake, as has occurred in lakes of North America and Scandinavia (Davis et al., 1985, Charifson et al., 2015). To disclose the absence of fish in Hältingträsk, further research on the history of fish populations in the lake is needed.

#### 6.4 Modern Times in Storträsk

The history of Lake Storträsk through the Megahlayan was described on account of the sediments physicochemical properties (trace metal concentrations, CNS analysis and LOI), and bioindicators (diatom, chironomid and cladoceran assemblages) in an earlier study (Rantala, 2013). The research concluded that Storträsk is naturally acidic and humic, and that the lake is sensitive to environmental stressors such as natural or human induced climate variability, modifications to nutrient input and atmospheric pollution (Rantala, 2013). However, low temporal resolution of the study did not allow estimation of potential signals of the  $\text{CaCO}_3$  treatments or the fish plantings. The purpose of this new revision is to study the resilience of Storträsk to these specific practices in recent years.

Although liming should have an effect on the water pH, only at two points (depths 3.5 and 2.5, Fig. 18), the reconstruction shows transient increase of pH values of ~6, which could be due to liming. But, without accurate records of  $\text{CaCO}_3$  treatment it is not possible to discern. In a similar fashion, the diatom assemblage does not show signals of change other than a decrease in *A. tenella* after the increase of pH (Fig.13 and 18). It has been referenced in past studies a change in the pH after liming treatment but without a recovery of the diatom community (Bishop et al., 2001, Renberg et al., 2009). The trends in Storträsk are contrary to previous research and displays resilience against liming.

While Storträsk appears to be adjusted to the liming treatment, the diatom assemblage suggests warmer temperatures may have an effect on the lake. The slight decrease of some *Aulacoseira* species such as *A. italic*, *A. lacustris* and *A. ambigua* in the top 2 cm of the record, paired with an increment of chlorophyll a (Fig. 13 and 26) can be attributed to climate warming. Similar trends, with decreasing *Aulacoseira* spp. has been assessed in Canadian and Swedish lakes (Shemesh et al., 2001, Harris et al., 2006). The drop of this genus has been associated to changes in the lake stratification, as *Aulacoseira* thrive in environments with higher turbulence, which may be reduced under warming (Harris et al., 2006, Rühland et al., 2015). Also, the species *A. formosa* and *Psammothidium marginulatum* are found in the sediments with low abundances. These species have been connected to climate warming in Canadian lakes (Sivarajah et al., 2016, Nelligan et al., 2016, Sivarajah et al., 2017). The cladoceran species in Storträsk had already given signs of affectations due to warming climate with the species *Bosmina longirostris* (Rantala,

2013). All in all, the bioindicators point out the sensitivity of Storträsk to the increment of temperature.

The reaction of algae communities after fish introduction has been researched recently in European and North American lakes (Charifson et al., 2015, Sienkiewicz and Gąsiorowski, 2016, Milardi et al., 2017). Charifson et al. (2015) and Sienkiewicz and Gąsiorowski (2016) arrived at the conclusion that the productivity of the lake is increased after the introduction of fish to the lake. While Sienkiewicz (2016) determined that the most intense changes happened after the fish introduction, Milardi and others (2017) only identified a change from planktonic to benthic diatoms. It is not possible to assure that the relative decrease of *A. tenella* starting from 1.25 cm is related to fish plantings, because the species still remains dominant. Nevertheless, the decrement of *Aulacoseira* the slight increment of fragilarioid diatoms in the top 2 cm (Fig. 13), and the higher values of chlorophyll a (Fig. 26) may allude to an increment in the productivity of the lake. Whereas these changes could be signs of the effects of fish introduction, more research on the nutrients input to the lake and its sources, as well as a historical record of the fish planting is needed.

### 6.5 Implications and Uncertainties

It is not possible to fully understand the variability and evolution of modern ecosystems only with environmental records, they do not go far back in time and therefore are not able to register the responses of ecosystems to different stressors. Lakes are sensitive to modifications in their catchment and the atmosphere, including changes such as erosion, vegetation change, and climatic variability that will have an effect on the properties of the lake. While natural changes usually translate into long term variation, current anthropogenic activities have caused fast-paced shifts of the physicochemical properties and biota of lakes. In this context, lakes in Sipoo which are naturally acidic and humic with *Sphagnum* bog in their surroundings, are susceptible to human activities such as agriculture, atmospheric pollution and climate change. With view of the future urban development in Östersundom, establish the reference conditions of Hältingträsk and assess the resilience to anthropogenic influences became a necessity. Studying the evolution of the lake through the mid-Holocene onwards allowed to understand Hältingträsk natural succession and detect past human influences. In addition, the high-

resolution core from Storträsk shed light on the little variation in the recent years and the small effects of liming and fish introduction.

Before being a lake, Hältingträsk was part of the Ancyclus Lake and later of the Litorina Sea. The present record was able to find typical diatoms of those stages, as well as evidence of the mineral deposition during these stages. During the Ancyclus regression the eroded material from the bedrock caused a high deposition of heavy metals, and the geochemistry of the Litorina Sea reflects similarities with the modern Baltic Sea, with high concentrations of metals, probably from manganese nodules. The isolation of Hältingträsk was a slow process between 7500 and 6500 cal BP that increased the organic compounds deposited and facilitated the presence of fragilarioid species and oscillations in the minerals fixated in the sediments.

The final stage of isolation took place between 6500 and 5000 cal BP, at this time the conditions surrounding the lake were warmer and drier than at present. The change of environment, from a marine to a fresh-water environment represented a change in biota and in the physicochemical properties. The records from Hältingträsk represent a common lake succession; being deeper and oligotrophic at the beginning and evolving along with the catchment. The different proxies were able to distinguish the formation of the peat bog surrounding the lake, as well as the effects on the water pH and biota. This process meant an acidification of the water and exhibit the large influence the vegetation has in the conditions of the lake. The pH reconstruction points out that the lake before intensification of agriculture was acidic due to natural acidification and particularly, the formation the *Sphagnum* vegetation around the lake, which provided humic substances to the water.

The lake also shows signs of sensitivity to erosion. For example, the period when slash-and-burn practices happened, is well represented in changes in the diatom community and variability of organic matter deposited. The presence of *A. formosa* is a notable sign of different environmental changes, this species increased with the intensification of agriculture, and decreased with the cooling period LIA, demonstrating its sensibility to not only nutrient enrichment, but also climate variability. On this regard, further analysis of the sediments would bring better explanations of the resilience of the lake, for example, analyzing the carbon to nitrogen (C/N) ratio or stable carbon isotopic ( $\delta^{13}\text{C}$ ) composition of organic matter would enable to distinguish the source of the organic matter.

The geochemistry of the sediments reveals Hältingträsk is responsive to atmospheric pollution as, it mirrors the evolution of European industrialization through the increment of heavy metals, in particular Pb. The water-pH is heavily influenced by the substances deposited in the lake. The increment of nutrients deposited in the catchment, caused by soil erosion from agriculture and other human activities, led to a process of alkalization in the early stages and later to the eutrophication of the lake. It is evident the relationship between the processes of alkalization or acidification with the different inputs leaking from the catchment (sulfur deposition from nutrient losses, acid runoff from ditching, and atmospheric pollution) still, to complement and confirm the progression of this events, further research on the cycles of P, N and S is needed.

Although an apparent recovery of the pH is perceived by the reconstruction and the modern records, the diatom community is different from the one found in the reference conditions, prior intensive agriculture and industrialization. This change is likely due to anthropogenic climate warming and the continuation of agricultural practices in the vicinity of the lake. The different shifts in the lake; variability of water pH and high trace metals concentrations after industrialization, could account for the lack of fish in Hältingträsk, nevertheless more research is required to conclude, especially more historical records regarding fish population.

Storträsk displays a partial recovery of the diatom assemblage, but it also shows the influence of climate warming in the decrease of *Aulacoseira* genus, the appearance of *A. Formosa* and *Psammothidium marginulatum* and an increment of Chlorophyll a. Regardless of the high resolution of the core, signals of possible affectations from fish plantings and liming are limited, there is only a small decrement of certain centric diatoms. Also, the productivity of the lake shows a slightly increasing trend. Nevertheless, to fully disclose the effects of these practices, a comparison of the changes in environmental variables with records of fish plantings and liming is recommended.

## 7. CONCLUSIONS

Lake Hältingträsk was part of the Ancylus Lake and the Litorina Sea until its isolation from the Baltic Basin. The transformation from glacial lake to fladmark was a slow process between 7500 and 6500 cal BP. The ontogeny of Hältingträsk suggests oligotrophic conditions at the beginning with slightly acidic waters, as the catchment evolved, the bog surrounding the lake was formed, changing the properties of the lake. According to the results,

Hälingträsk is a naturally acidic lake influenced by the nearby vegetation and sensitive to soil erosion.

As southern Finland was populated, the signs of early agriculture can be track in the lake sediments, proving Hälingträsk is susceptible to anthropogenic pressures in the catchment. In this regard, shifts in *A. formosa* outlines periods of increasing nutrient input, as well as warmer periods. The European industrialization of the late 1800s onwards is reflected in the geochemistry of the sediments, showing that Hälingträsk is affected by atmospheric contamination. Additionally, mechanisms such as discharge of substances, loss of acid sulfate soils, agriculture and weak acid rain will translate into processes of alkalization or acidification, changing the water pH. Thus, Lake Hälingträsk is sensitive to modifications in its catchment due to soil erosion and leaking of nutrients, its chemistry can be altered by atmospheric pollution, and both natural and anthropogenic climate change.

The high resolution study of the most recent years from Storträsk suggests the lake is resilient against certain stressors such as liming and fish introduction, showing faint signals of both. However, detailed historical records of these practices are required to conclude. In contrast, the diatom assemblage shifts may further have been affected by the recent warming. In summary, few changes are registered in the sediments from the past 10 years.

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